

PATENT COOPERATION TREATY

PCT

NOTIFICATION OF ELECTION

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

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in its capacity as elected Office

Date of mailing (day/month/year)

28 August 2000 (28.08.00)

International application No.

PCT/AU00/00018

Applicant's or agent's file reference

IRN 606621

International filing date (day/month/year)

17 January 2000 (17.01.00)

Priority date (day/month/year)

18 January 1999 (18.01.99)

Applicant

SIVASITHAMBARAM PILLAI MAILVAGANAM PILLAI, Mailvaganam, Thavalingam et al

1. The designated Office is hereby notified of its election made:



in the demand filed with the International Preliminary Examining Authority on:

26 June 2000 (26.06.00)



in a notice effecting later election filed with the International Bureau on:

2. The election ☒ was

was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

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(19) World Intellectual Property Organization
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C08K 3/36

[MY/MY]; Suite 101, 1st floor, Wisma Kwang Tung Association Building, 44 Jalan Pudu, 55100 Kuala Lumpur (MY).

(21) International Application Number: PCT/AU00/00018

(74) Agent: **PHILLIPS ORMONDE & FITZPATRICK**; 367 Collins Street, Melbourne, Victoria 3000 (AU).

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For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: USE OF VEGETATIVE MATERIAL AS A FILLER IN COMPOSITE MATERIALS

(57) Abstract: The present invention provides a filler for use in composite materials wherein said filler comprises a vegetative-based material and wherein said vegetative-based material can be fresh or carbonised. In one particularly preferred embodiment the present invention utilises carbonised rice husk. In a further aspect of the present invention there is provided a process for the production of a carbonised vegetative-based filler wherein said process comprises burning a fresh vegetative-based material at about 800 °C for about 4 seconds.



WO 00/042116 A3

INTERNATIONAL SEARCH REPORT

International application No.

PCT/AU 00/00018

A. CLASSIFICATION OF SUBJECT MATTER												
Int Cl ⁷ : C08L 97/02 C08K 3/36												
According to International Patent Classification (IPC) or to both national classification and IPC												
B. FIELDS SEARCHED												
Minimum documentation searched (classification system followed by classification symbols) IPC C08K C08L (together with keywords as stated below)												
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched AU: IPC as above												
Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) WPAT : rice AND (husk OR hull) AND (carboni: OR bitumen OR resin OR rubber OR ebonite OR concrete) AND IPC as above. JAPIO : as for WPAT												
C. DOCUMENTS CONSIDERED TO BE RELEVANT												
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.										
X	AU 68923/94 A (PAGDEN) 13 July 1995 Page 4	1,2										
X	AU 60599/94 A (NATIONAL RESEARCH DEVELOPMENT CORPORATION) 2 November 1995 Examples 1-6	1,2										
X	AU 34510/95 A (PAGDEN) 1 February 1996 Examples 1-6	1,2,33										
<input checked="" type="checkbox"/> Further documents are listed in the continuation of Box C <input checked="" type="checkbox"/> See patent family annex												
<p>* Special categories of cited documents:</p> <table border="0"> <tr> <td>"A" document defining the general state of the art which is not considered to be of particular relevance</td> <td>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</td> </tr> <tr> <td>"E" earlier application or patent but published on or after the international filing date</td> <td>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone</td> </tr> <tr> <td>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</td> <td>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art</td> </tr> <tr> <td>"O" document referring to an oral disclosure, use, exhibition or other means</td> <td>"&" document member of the same patent family</td> </tr> <tr> <td>"P" document published prior to the international filing date but later than the priority date claimed</td> <td></td> </tr> </table>			"A" document defining the general state of the art which is not considered to be of particular relevance	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention	"E" earlier application or patent but published on or after the international filing date	"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone	"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art	"O" document referring to an oral disclosure, use, exhibition or other means	"&" document member of the same patent family	"P" document published prior to the international filing date but later than the priority date claimed	
"A" document defining the general state of the art which is not considered to be of particular relevance	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention											
"E" earlier application or patent but published on or after the international filing date	"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone											
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art											
"O" document referring to an oral disclosure, use, exhibition or other means	"&" document member of the same patent family											
"P" document published prior to the international filing date but later than the priority date claimed												
Date of the actual completion of the international search 21 February 2000		Date of mailing of the international search report 28 FEB 2000										
Name and mailing address of the ISA/AU AUSTRALIAN PATENT OFFICE PO BOX 200, WODEN ACT 2606, AUSTRALIA E-mail address: pct@ipaustalia.gov.au Facsimile No. (02) 6285 3929		Authorized officer N.L. KING Telephone No.: (02) 6283 2150										

INTERNATIONAL SEARCH REPORT

International application No.

PCT/AU 00/00018

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	GB 2084212 A (ALLAMI EPITOIPARI VALLALAT) 7 April 1982 Examples 1-3	1,2
X	JP 4045156 A (YASUTAKU) 14 February 1992 Derwent Abstract 92-108756	1,2
X	JP 1249617 A (DENKI KAGAKU KOGYO KK) 4 October 1989 Derwent Abstract 89-335790	1-13
X	US 3988270 A (WILLIS et al) 26 October 1976 Examples 1-4	1,2,23,24
X	SU 956502 A (MELKUMOV) 7 September 1982 Derwent Abstract 83-715109	1,2,21
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X	JP 55139441 A (MATSUSHITA ELECTRIC IND CO LTD) 31 October 1980 JAPIO Abstract	1-4,14,17,18, 37
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X	BE 1000901 A (DE NEEF) 9 May 1989 Derwent Abstract 89-145267	1,2,39
X	US 4105459 A (MEHTA) 8 August 1978 Examples 1-25	1-5,14,15, 17,18,39
X	WO 92/00251 A (MEHTA) 9 January 1992 Examples 1-8	1-5,14,15, 17,18,39,40

**AUSTRALIAN PATENT OFFICE
SEARCH REPORT**

International application No.
PCT/AU 00/00018

C (Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 3951907 A (MEHTA) 20 April 1976 Examples 1-22	1-5,14-18,23, 24,27,28
X	GB 955049 A (GULF STATES ASPHALT COMPANY INC.) 8 April 1964 Pages 3,4	1,2,19,20

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/AU 00/00018

This Annex lists the known "A" publication level patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

Patent Document Cited in Search Report				Patent Family Member			
GB	2084212	AT	3941/81	CH	653078	DE	3136521
		ES	506045	FR	2490142	GR	75084
		IN	154800	IT	1139168		
WO	9521550	AU	15767/95	IT	1273768		
US	4202803	AR	219752	AU	37115/78	BR	7803885
		DE	2823872	ES	470627	FR	2395287
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		GB	2149807	JP	60124641	NL	8403329
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		DE	2338066	ES	417347	FR	2194760
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		JP	49132108	NL	7310493	PH	11658
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		HK	1001173	HU	67417	NO	924945
		US	5346548				
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		BR	7305735	DE	2338066	ES	417347
		FR	2194760	GB	1432707	IN	138985
		IN	140364	IN	142045	IT	1006067

International application No.
PCT/AU 00/00018

NL

7310493

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11658

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4105459

END OF ANNEX

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PATENT COOPERATION TREATY

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- 3 JUL 2000

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INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

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MELBOURNE VIC 3000

NOTIFICATION OF RECEIPT
OF DEMAND BY COMPETENT INTERNATIONAL
PRELIMINARY EXAMINING AUTHORITY

(PCT Rule 59.3(e) and 61.1(b), first sentence
and Administrative Instructions, Section 601(a))

Date of mailing 30 JUN 2000
(day/month/year) (30/6/00)

Applicant's or agent's file reference
IRN606621

IMPORTANT NOTIFICATION

International application No.
PCT/AU00/00018

International filing date (day/month/year)
17 JAN 2000 (17/1/00)

Priority date (day/month/year)
18 JAN 1999 (18/1/99)

Applicant

Contract Research & Development (M) Sdn. Bhd. (et al.)

1. The applicant is hereby **notified** that this International Preliminary Examining Authority considers the following date as the date of receipt of the demand for international preliminary examination of the international application:

26 JUN 2000 (26/6/00)

2. That date of receipt is:



the actual date of receipt of the demand by this Authority (Rule 61.1(b)).



the actual date of receipt of the demand on behalf of this Authority (Rule 59.3(e)).



the date on which this Authority has, in response to the Invitation to correct defects in the demand (Form PCT/IPEA/404), received the required corrections.

3. ☐ **Attention:** That date of receipt is **AFTER** the expiration of 19 months from the priority date. Consequently, the elections(s) made in the demand does (do) not have the effect of postponing the entry into the national phase until 30 months from the priority date (or later in some Offices) (Article 39(1)). Therefore, the acts for entry into the national phase must be performed within 20 months from the priority date (or later in some Offices) (Article 22). For details, see the *PCT Applicant's Guide, Volume II*.



(If applicable) This notification confirms the information given by telephone, facsimile transmission or in person on:

4. Only where paragraph 3 applies, a copy of this notification has been sent to the International Bureau.

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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁷ : C09K	A2	(11) International Publication Number: WO 00/42116 (43) International Publication Date: 20 July 2000 (20.07.00)
(21) International Application Number: PCT/AU00/00018 (22) International Filing Date: 17 January 2000 (17.01.00) (30) Priority Data: PP 8198 18 January 1999 (18.01.99) AU (71) Applicants (for all designated States except US): CONTRACT RESEARCH & DEVELOPMENT (M) SDN. BHD. [MY/MY]; Suite 101, 1st floor, Wisma Kwang Tung Association Building, 44 Jalan Pudu, 55100 Kuala Lumpur (MY). TADGELL, David, John [AU/AU]; 367 Collins Street, Melbourne, Victoria 3000 (AU). (72) Inventors; and (75) Inventors/Applicants (for US only): SIVASITHAMBARAM PILLAI MAILVAGANAM PILLAI, Mailvaganam, Thavalingam [MY/MY]; (MY). HAJI AHMAD, Khalid [MY/MY]; (MY). VETTIVALOO ARUNASALAM, Arulgnanam [MY/MY]; Suite 101, 1st floor, Wisma Kwang Tung Association Building, 44 Jalan Pudu, 55100 Kuala Lumpur (MY). (74) Agent: PHILLIPS ORMONDE & FITZPATRICK; 367 Collins Street, Melbourne, Victoria 3000 (AU).		(81) Designated States: AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG). Published <i>Without international search report and to be republished upon receipt of that report.</i>
(54) Title: USE OF VEGETATIVE MATERIAL AS A FILLER IN COMPOSITE MATERIALS (57) Abstract The present invention provides a filler for use in composite materials wherein said filler comprises a vegetative-based material and wherein said vegetative-based material can be fresh or carbonised. In one particularly preferred embodiment the present invention utilises carbonised rice husk. In a further aspect of the present invention there is provided a process for the production of a carbonised vegetative-based filler wherein said process comprises burning a fresh vegetative-based material at about 800 °C for about 4 seconds.		

USE OF VEGETATIVE MATERIAL AS A FILLER IN COMPOSITE MATERIALS

TECHNICAL FIELD

5 This invention relates to the use of a filler derived from cereal husk, more particularly rice husk, in composite materials to enhance the flame retardant, antistatic, accelerator, plasticiser and blowing characteristics in various composite materials. The invention has particular but not exclusive application to the following families of composites:-

- 10 1. Thermoplastic Resins
2. Thermoset Plastics
3. Rubbers and Elastomeric Materials
4. Conductive Coatings and Printing Inks
5. Bitumen
- 15 6. Concrete

BACKGROUND ART

Composite materials are well known. Fillers are usually added to composite materials, including composite polymers, to save cost or to enhance a particular mechanical property or other characteristic of the materials. The usage of fillers is usually accompanied by coupling agents that enhance the polymer-filler and filler-filler interaction so that the expected properties are realised.

The present invention is concerned with fillers which enhance the anti-static, flame retardant, accelerator, plasticiser, blowing characteristic and/or other physical or mechanical properties of composite materials and has particular application for use in composite polymers. Such have wide application in the aeronautical, mining, computer, road building, textile, foot ware, rubber and polyurethane industries among others. For example, it is often desirable to prevent the build up of static charges which can cause sparks (and hence explosions or electrical damage) or production problems, eg. collection of dust and poor feeding of materials through machinery. More highly conductive composite polymers can also be used for Electro Magnetic Interference shielding, for example.

Carbon black fillers, aluminium flakes and fibres, stainless steel-fibres and chopped carbon fibres have all been used as fillers for the purpose of rendering composite plastic conductive. Likewise other chemicals such as Halogen compounds or triethyl phosphate have been used to achieve the flame retardant property.

A number of theories have been proposed to explain how discreet particle fillers impart conductivity and flame retardant properties in composite plastics.

In order for current to flow in a conductive polymer compound, electrons must travel along the filler as the plastic itself is an excellent insulator. To achieve this flow the discreet particles of the filler must be in contact or separate by a minimum distance which is probably less than 100 Angstroms. There are three properties of the filler particles which will effect the average inter-particle distance for a given filler loading in a polymer system. These are particle size, shape (structure), and porosity. Smaller size, irregular shape and high porosity all result in smaller inter-particle distances and hence higher conductivity. A fourth property of the particle which is relevant to conductivity and flame retardant properties in the composite plastics is surface chemistry, that is the presence of oxygen on the surface. The presence of appreciable quantities of oxygen on the surface (called volatile content) acts as insulation and hence reduces conductivity.

The known conductive fillers such as carbon black, aluminium, stainless steel and carbon fibres are expensive and furthermore some of these materials have other processing difficulties, eg. aluminium fibres and stainless steel fibres settle in liquid environments due to their high density. Further problems with known conductive fillers are that they often compromise other properties of composite plastics such as flame retardance and strength.

Static electrification of articles can lead to a number of undesirable effects including:

- Attraction of dust particles.
- Attraction between surfaces, e.g. plastic films and textile yarns.
- Risk of fire or explosion caused by sparking near inflammable liquids, gases, and explosive dusts, e.g. coal dust and flour.
- Risk of shock to persons handling equipment.

The accumulation of electrostatic charges can be prevented by using materials of low resistance. The resistivity of natural rubber can be lowered by compounding with suitable ingredients. Alternatively, as static electrification is a surface phenomenon, the product can be covered with a conducting surface layer.

Low resistance rubber is required for a wide range of applications, such as rollers for textile machinery, conveyor belting, fuel hoses, flooring, footwear, antistatic gloves (electronic industry), cables, equipment used in hospital operating theatres, and aircraft components.

The terms "antistatic" and "conductive" are restricted here to rubber products rather than the rubber itself because the electrical resistance of the product depends not only on the resistivity of the rubber but also on the shape and most probable positions of charge generation and discharge.

Natural rubber is normally considered to be an electrically insulating material but it can be an electrically insulating material but it can be compounded to give electrical resistivity lying anywhere between 1 ohm/cm. and 10^{15} ohm/cm. The most common means of reducing resistance is to add a suitable carbon black (super conductive furnace). Resistance falls with a decrease in particle size, increase in black "structure" and increase in concentration. For light coloured products certain grades of aluminium silicate may be used as antistatic fillers although these are usually less effective in reducing resistance than the super conductive furnace. There are also other proprietary antistatic agents that are available, such as ethylene oxide, but still these agents are less effective than the super conductive furnace.

The applicant has found that cereal husk (more preferably but not exclusively carbonised rice husk) is particularly suited for use as a filler in plastics as it has been found to enhance the conductivity and flame retardant properties of the composite plastics. The presence of appreciable quantities of oxygen on the surface of carbonised rice husk acts as insulation for each aggregate, thereby reducing the conductivity and also reducing the flammability. The presence of nitrogen and oxygen in the fresh husk not only enables the blowing effect but also nitrogen being inert reduces the flame spread. The volume of gas (nitrogen/oxygen) evolution per gram of fresh rice husk is 240 ml/g. The husk's

decomposition temperature is at about 280°C and curing temperature of rubber and ethyl vinyl acetate is between 130°C-180°C, thus when urea is milled along the decomposition temperature is reduced within the curing temperatures. The presence of silica in the rice husk provides better mechanical strength.

5 Typical chemical and physical properties of fresh and carbonised rice husk are detailed as follows:

- consists of 20-23% of paddy
- husk burning: 20% ash by weight
90-95% is silica (amorphous and crystalline)
- 10 • physical characteristics: bulk density 96.12-112.14 kg/m³
- pH 7.14 (husk ash)
- moisture content 5.6-7.2%, dry basis
- ash 22.2%

15 Chemical Composition

	Moisture Content	: 5.6-7.2%, dry basis
	Ash:	: 22.2%
	Protein	: 2.4%
	Crude fat	: 0.7%
20	Carbohydrate	: 32.0%

		Fresh RH	Carbonised RH
	Al ₂ O ₃	0.025%	0.023%
	CaO	0.36%	0.12%
25	NaO	0.034%	0.018%
	SiO ₂	96.2%	53.88%
	Fe ₂ O ₃	0.041%	0.022%
	MgO	0.16%	0.078%
	K ₂ O	0.69%	0.95%
30	P ₂ O ₅	0.57%	0.27%

It is an object according to one aspect of the present invention to provide an alternative filler which will enhance the antistatic, flame retardant, accelerator, plasticiser blowing and/or other physical or mechanical in composite materials.

35 The filler is desirably cheap, environmentally friendly and replenishable and it does not compromise other characteristics of the composite material.

SUMMARY OF INVENTION

The present invention in one aspect resides in the use of fresh and/or carbonised vegetative-based filler to provide improved antistatic, flame retardant, accelerator, plasticiser, blowing and/or other physical or mechanical characteristics in composite materials.

Preferably, the fresh vegetative-based filler is fresh/raw rice husk. Fresh rice husk is obtained after the husk is separated from the grain. The husk is ground by any mechanical process, i.e., using hammer mill or cryogenic or jet mill. As the fresh rice husk is used in various composite materials to determine various properties therefore it is imperative to determine the particle size for each usage. For example with bitumen a particle size of 100 mesh size, blowing agent a particle size of 325-400 mesh size, antistatic and flame retardant in thermoset a particle size of 325 mesh size.

Preferably, the carbonised vegetative-based filler is carbonised rice husk.

Preferably, the carbonised rice husk is burnt at about 800°C for about 4 seconds. Most preferably, the carbonised rice husk is burnt at 803-804°C for 3-4 seconds.

In another aspect the invention resides in a composite material, more particularly a composite plastic including a vegetative-based filler when used as a conductive or flame retardant article.

Preferably, the carbonised vegetative-based filler is carbonised rice husk which has been burnt at 803-804°C for 3-4 seconds.

The present invention also exhibits the usage of fresh and carbonised rice husk as a blowing agent when used in combination with recycled (reclaimed), or virgin natural rubber or other suitable thermoplastic materials. Though other conventional blowing agents have been used with natural or synthetic rubber to achieve the similar products but so far no blowing agents have been used with recycle (reclaim) rubber to produce similar products. Furthermore the conventional blowing agents are expensive and dosages are higher as compared to the fresh rice husk. For example for the conventional blowing agent, the dosage is about 6.5-7 phr, whereas the fresh/carbonised rice husk, the dosage is between 1.5 to 3 phr. When rice husk is used at different dosages the blowing effect is different. It was also noted that the rice husk does not only work as a

blowing agent, but also as a plasticizer and a filler. The properties achieved are comparable to conventional blowing agents, when using fresh or carbonised rice, has no difference to the conventional blowing agent other than the colour of the end product.

5 Ebonite, a hard, dark-coloured plastic-like material, is the reaction product of rubber and a large proportion of sulphur. Simple rubber / sulphur mixtures are seldom used in practice; they suffer from poor processability, require long cure times and lead to excessive shrinkage and heat evolution during cure. Accelerators, fillers, processing aids and other compounding ingredients are
10 widely used in ebonite, as in soft rubber vulcanised rubber, to ease processing, shorten cure times and modify properties. The curing times for ebonite are generally up to ten (10) hours at 150° C, thus making ebonite products expensive. Ebonite can be made from synthetic, such as BR, NBR, SBR and Nitrile rubber and as well as from Natural rubber. High strength, low thermal conductivity,
15 chemical resistance and insulating properties of natural rubber make it a popular choice. Although it has been superseded in many applications by synthetic thermoplastic and thermosets, it is still used for outstanding chemical resistance and electrical properties coupled with high mechanical strength and ease of machining.

20 The present invention exhibits the usage of fresh and carbonised rice as an accelerator when used in combination with recycled (devulcanised) or virgin natural rubber, and at the same time making ebonite a conductive product when carbonised rice husk is used. Though other conventional accelerators have been used with natural or synthetic rubber (virgin or recycled) to achieve the similar
25 products but so far no accelerators like the rice husk material have been used with recycled (devulcanised) rubber to produce similar products. Further more the conventional accelerators and conductive carbon black are expensive and difficult to blend and process. When rice husk is used singularly at different dosages the activation effect is different to meet industrial requirements.
30 Generally for ebonite production the sulphur content should be in the range of 25-40 phr, but whereas when fresh rice husk between 25-30 phr is used the sulphur content could be reduced to 20-25 phr. Accelerators are less effective in ebonite than in soft rubber and large quantities are generally required. Basic accelerators

such as guanidines and aldehyde-amines are preferred. Inorganic activators such as magnesium oxide, magnesium carbonate and lime are also effective when used with organic accelerators to reduce cure time without the risk of over heating.

5 Common inorganic fillers used in ebonite are china clay, talc, silica, whiting and magnesium oxide. These also reduce shrinkage and heat evolution but loaded ebonite generally have weaker mechanical properties than unloaded ones. Carbon black does not reinforce ebonite and is normally only added for pigmentation, although conductive carbon black are sometimes used to prepare
10 electrically conducting ebonite.

BEST MODE

Following is an example of the invention, in this example the filler is carbonised rice husk (CRH) which has been burnt at 803-804°C for 3-4 seconds.
15 After this the CRH is obtained.

The manner in which the rice husk is burnt is believed to be important in achieving the desired surface area, surface structure and porosity necessary for conductivity and flame retardant and blowing properties in the composite plastics to be achieved. At this stage the range of temperature and the duration of the
20 time of burning which achieves the desired result has not been fully explored, however it is predicted that the temperature range will be from about 100-950°C and the time range will be from about 2-30 seconds, although these ranges may be much narrower. The importance of controlled burning in a prescribed time results in obtaining better surface area and porosity which in turn offers ideal
25 properties emitting anti-static, flame retardant and enhancing physical properties of the material. In the absence of controlled burning, the surface area, surface structure and porosity would be distorted. While the present exemplification involves use of carbonised rice husk it is possible that the desired results may be achieved by use of other carbonised vegetative-based fillers.

30 EXAMPLE 1

A thermoset application called pulforming was used to manufacture fibre reinforced bolts. Fibre glass tows (36 tow of 8000 tex) are pulled into a resin bath that contains:

1. Polyester and Vinyl Ester combination, ie. 60% Vinyl Ester (Derakane 411 - Dow Chemical) and 40% Polyester (Everpol 3260 AR - P.T. Arinde).
2. Zinc Stearate (mould releasing agent) - 1.18% of the resin weight
BYK 980 (improves wetting and dispersing of fillers in glass fibre reinforcement compounds) - 1.5% of the filler weight.
3. BYK 515 (air releasing agent) - 0.5% of the total weight of the resin mixture.
4. BYK 996 (wetting and dispersing additive for mineral fillers in hot curing, glass fibre reinforced UP-resin systems) - 2% of the resin weight.
5. Fillers ($\text{Ca}_2(\text{CO})_3$ & Carbonised Rice Husk (mesh size 325) @ 55% & 12% of the resin weight).
6. Aluminium Trihydrate (2.4% of the resin weight).
7. Catalyst TBPH (Tertiary Butyl Peroxy-2-Ethyl Hexanoate) - 2.12% of the resin weight.
TBPB (Tertiary Butyl Perbenzoate) - 0.53% of the resin weight.

The wet fibre glass tow is pulled into the mould and compressed at a pressure of 800psi (5600kPa) for 3.8 minutes at 130°C. Then the bolt is pulled out of the mould and left to cure.

The following day tests were carried out on the bolt with the following results:

Tensile strength at the thread - 50kN

Torque - 45ft/lb

Bond strength - BS 1610:Part 1, Grade 1.0

Fire rating - BS 5865:1980 - Persistence flame shall be less than 10 seconds

Electrical conductivity - less than 10 to the power of 9 Ohms.

EXAMPLE 2

All chemicals used are taken by percentage of weight of rubber. The rubber and the chemicals are mixed in a Banbury, for 5 minutes. Recycled rubber

(reclaim) (220g) is first milled with zinc oxide (4.5%) - accelerator, which is followed with stearic acid (1.8%) - activator, Mercadibenzothiazole disulphide (MBTS)(0.5%), Tetramethylthiuram disulphide (TMTD)(0.2%)- accelerator, fresh rice husk (27%)-blowing agent and filler and sulphur (2.7%)- vulcanisate. Then the mixed compound is milled for five (5) minutes to form a sheet that is ready for curing. Then a piece of the sheet weighing about 32g is placed in a mould that it is to be cured for two (2) minutes in a oven at 150°C temperature. The conventional curing time is six (6) minutes at the same temperature of 150°C.

The rubber and the chemicals are mixed in a Banbury, for 5 minutes. The similar approach has been done for using SBR Rubber (100g), silica (62g), Peg 1500 (2.5g), Paraffin oil (5g), Zinc oxide (2.5g), Wing stay (0.5g), Wax (1g), Mercadibenzothiazole disulphide (MBTS) (1.5g), Tetramethylthiuram disulphide (TMTD) (0.2g), Stearic acid (1.5g) and Sulphur (2g). The milling was done for ten (10) minutes and later cured in the oven for six (6) minutes at 150°C.

This exercise was repeated by using fifty (50) percent of the virgin material compound and fifty (50) percent recycled (reclaimed) material compound, and cured in the oven at 150°C for two (2) minutes.

With the level, of rice husk dosage, the blowing effect can be controlled to suit the industry's requirement.

Machine Operating Conditions

Virgin Rubber:

Mix properties; White filled mix

Mooney viscosity, MLI + 3, 100°C	24
Mooney viscosity, MLI + 3, 120°C	18.5
Mooney scorch, t ₅ MLI + 3, 120°C min	5.8
Monsanto Rheometer, 160°C	
time to 95% cross-linking, s	110

By using rice husk the Mooney viscosity was lower than the conventional filler, thus lowering scotch time (time taken by the rubber compound to flow into the

mould) and curing time (time taken to cure rubber compound) respectively. As such this leads to a cheaper production system. Presently various fillers and blowing agents are being used in the production of soft/spongy rubber that would produce different types of cell structures for an end product, but the cost
5 determines the market.

EXAMPLE 3

All chemicals used are taken by percentage of weight of rubber. The rubber and the chemicals are mixed in a Banbury, for six (6) minutes. The
10 recycle (devulcanised) rubber is first milled with magnesium oxide (2%) - accelerator, which is followed with Diphenylguanidine (2%) - accelerator, fresh rice husk (30%) - accelerator and filler and sulphur (30%) - vulcanisate. After the milling at the Banbury for ten (10) minutes, it is then milled into a sheet. The mould was heated in the oven press to 150°C then the sheeted rubber is placed
15 in the mould and it is cured for twelve (12) minutes. The conventional curing time is between eight to ten hours at the same temperature of 150°C.

A conventional formula for ebonite was selected to compare. The rubber and the chemicals are mixed in a Banbury, for 5 minutes. The similar mixing as above was followed, using SBR 5 Rubber (100g), ebonite dust (100g), China clay
20 (50g), Magnesium oxide (5g), Diphenylguanidine (3g), Linseed oil (5g) and Sulphur (45g). The milling was done for ten (10) minutes and later cured in the oven for eight (8) hours at 150°C.

Mix properties; Rice Husk filled mix

25	Mooney viscosity, MLI + 3, 100°C	24
	Mooney viscosity, MLI + 3, 120°C	18.5
	Mooney scorch, t_5 , MLI + 3, 120°C min.	5.8
	Monsanto Rheometer, 160°C	
	time to 95% cross-linking, s	110

By using rice husk the curing time is reduced tremendously twelve minutes as compared to eight to ten hours. The sulphur content in the rubber polymer is reduced by fifteen percent.

5 EXAMPLE 4

All chemicals used are taken by percentage of weight of natural rubber (NR). The natural rubber and the chemicals are mixed in an open mill or kinder, for six (6) minutes. Natural rubber is first milled with stearic acid (1%) and zinc oxide (5%) activator, which is followed with rice husk (blowing agent) (2.5-3.5%), calcium carbonate - (40%), promoter - urea based (2.5-3.5%), silica (10%), accelerator dibenzthiazyldisulphide (MBTS) (0.05%) and catalyst sulphur (1.5%). After the milling at the open mill or kinder for ten (10) minutes, it is then milled into a sheet. The mould was heated in the oven press to 160°C then the sheeted natural rubber is placed in the mould and it is cured for twenty-two (22) minutes.

The temperature for curing could be from 145°-160°C and the cure time may differ according to the mould size.

Cured properties; rice husk filled blowed mix - micro-cellular cells.

	1.	Hardness	Askar C	35
20	2.	Shrinkage	%	5
	3.	Specific Gravity	g/cc	0.3-0.35

By using rice husk as a blowing agent the catalyst percentage could be reduced and as well as the percentage of blowing agent used.

25

EXAMPLE 5

THERMOPLASTIC (EVA)

All chemicals used are taken by percentage of weight of thermoplastic - Ethyl Vinyl Acetate (EVA). The EVA and the chemicals are mixed in an Open Mill or Knider, for six (6) minutes. Ethyl Vinyl Acetate (EVA) is first milled with Stearic Acid (1%) and zinc oxide (5%) accelerator, which is followed with Rice Husk (Blowing agent) (2.5%). Magnesium carbonate - (10%), Promoter - urea based (5%) and catalyst Diacyl Peroxide (1%). After the milling at the Open mill or

Knider for ten (10) minutes, it is then milled into a sheet. The mould was heated in the oven press to 160°C then the sheeted EVA is placed in the mould and it is cured for twenty-two (22) minutes.

5 The temperature for curing could be from 145°-160°C and the cure time may differ according to the mould size.

Cured properties: Rice Husk filled blowed mix - micro-cellular cells

1.	HARDNESS	Askar C	29-35
2.	SHRINKAGE	%	2
3.	SPECIFIC GRAVITY	g/cc	0.200
10 4.	COMPRESSION SET	%	80

By using rice husk as a blowing agent the catalyst percentage could be reduced and as well as the percentage of blowing agent used.

15 **EXAMPLE 6**

The rice husk is mixed by weight with tyre crumbs(35-40 mesh) and an effluent from the palm oil mill called Scavenger (which have a fatty acid content (C₈-C₁₈). From literature it has been reported that by using tyre crumb with the binder (bitumen) there is an improve of properties for the asphalt mixture. This
20 invention not only uses tyre crumb along with rice husk and an oil palm effluent to further improve the properties. As well as the formulation address the recyclability of all agro waste by-products to be used in the road surfacing industries. The formulation of the rice husk mixture as follows:

25	Rice husk	50%
	Tyre crumb	45%
	Scavenger	5%

In this particular example the usage of rice husk mixture is divided into two categories:

- 30 A. RICE HUSK MIXTURE USED IN MODIFIED BINDER
B. RICE HUSK MIXTURE USED IN AS FILLER

- 5 A. The rice husk mixture is added to the bitumen first in compliance to the SOCIETY OF HIGHWAY PROCEDURE (SHRP) to manufacture modified bitumen. The bitumen is first heated to about 160°C, then the rice husk mixture twenty percent 20% by weight of bitumen is mixed with the heated bitumen for about one hour. As a result of this reaction a modified bitumen is made. From here 5-7% by weight of this modified bitumen is added to the aggregate. The aggregate is first heated to about 200°C and the modified bitumen is mixed for three to four minutes. The modified bitumen with rice husk mixture complies to all requirement of the SHRP.
- 10 B. The rice husk mixture is added as a filler to the aggregate, by four (4%) by weight to the aggregate weight. The aggregate is first heated to 200°C, and is allowed to cool to about 160°C, then the rice husk mixture is added and mixed and lastly the bitumen 5-6% by weight of aggregate is added and mixed for 3-4 minutes. This blending with rice husk mixture complies to all requirement of the Marshall Properties.
- 15

TABLE 1

PROPERTIES OF RICE HUSK MODIFIED BINDER - SHRP

SHRP	80/100	RICE HUSK MIXTURE
FLASH POINT TEMP. °C	230	240
SOFTENING POINT, °C	44-50	55-70
PENETRATION @ °C 25, dmm	80-100	60-100
BROOKFIELD VISCOSITY @ 135°C, MPaS	<500	>1500
DYNAMIC SHEAR RHEOMETER		
PG 70		
ORIGINAL		
G* (Pa)	<1000	>1200
$\delta(^{\circ})$	>80	>80
G*/Sin δ	<1000	>1200
AFTER RTFOT		
G* (Pa)	<1000	>3800
$\delta(^{\circ})$	>80	>70
G*/Sin δ	<1000	>3800
AFTER PAV		
G* (Pa)	<1000	>230
$\delta(^{\circ})$	>80	>50
G*/Sin δ	<1000	>260
PG 76		
ORIGINAL		
G* (Pa)	<1000	>1800
$\delta(^{\circ})$	>80	>70
G*/Sin δ	<1000	>1800
AFTER RTFOT		
G* (Pa)	<1000	>2400
$\delta(^{\circ})$	>80	>70
G*/Sin δ	<1000	>2600
AFTER PAV		
G* (Pa)	<1000	>230
$\delta(^{\circ})$	>80	>50
G*/Sin δ	<1000	>280

TABLE 2

MIXED PROPERTIES OF RICE HUSK MODIFIED BINDER

PROPERTIES	80/100	RICE HUSK MIXTURE
MARSHALL STABILITY (kN)	5-10	>13
FLOW (mm)	2-4	2-4
QUOTIENT (kN/mm)	1-3.5	3-4
RESILIENT MODULUS @ 25°	>2000	>2800

TABLE 3

MIXED PROPERTIES OF RICE HUSK MIXTURE AS FILLER

PROPERTIES	80/100	RICE HUSK MIXTURE
MARSHALL STABILITY (kN)	6-10	>12
FLOW (mm)	2-4	2-4
QUOTIENT (kN/mm)	1-3.5	3-4
RESILIENT MODULUS @ 25°	>2000	>2800

TABLE 4

PREFERRED PARTICLE SIZE AND DOSAGE OF FRESH AND/OR CARBONISED RICE HUSK FOR PARTICULAR COMPOSITE MATERIALS

COMPOSITE MATERIAL	FRESH/DOSAGE	CARBONISED/DOSAGE
BITUMEN (MECHANICAL PROPERTY)	100 MESH - 40-60 phr	-
THERMOPLASTIC (EVA) BLOWING CHARACTER	325-400 MESH - 1.5-2.5 phr	325-400 MESH - 1.5-2.6 phr
THERMOPLASTIC (EVA) MECHANICAL PROPERTY	325-400 MESH - 1.5-5 phr	325-400 MESH - 1.5-2.5 phr
RUBBER (N.R./S.R.) BLOWING CHARACTER	325-400 MESH - 1.5-27 phr	325-400 MESH - 1.5-27 phr
EBONITE N.R. (REDUCE CURE TIME)	100-200 MESH 18-30 phr	-
RUBBER (N.R./S.R.) SCOTCH TIME	100-200 MESH 5-10 phr	100-200 MESH 5-10 phr
THERMOSET RESIN (FLAME PROPERTY)	-	325 MESH 10-15 phr
THERMOSET RESIN (MECHANICAL PROPERTY)	100-200 MESH 10-15 phr	100-200 MESH 10-15 phr
THERMOSET RESIN (ANTISTATIC)	-	325 MESH 10-15 phr
RUBBER-LATEX (N.R./S.R.) ANTISTATIC	-	325-400 MESH 5-15 phr
RUBBER (N.R./S.R.) ANTISTATIC	-	325-400 MESH 5-15 phr
CONCRETE (MECHANICAL PROPERTY)	100-200 MESH 10-15 phr	100-200 MESH 10-15 phr

N.R. - NATURAL RUBBER
S.R. - SYNTHETIC RUBBER

10 It will of course be realised that whilst the above has been given by way of illustrative examples of this invention, all such and other modifications and variations hereto, as would be apparent to persons skilled in the art, are deemed to fall within the broad scope and ambit of this invention as herein set forth. For instance, while the preceding examples relate to the use of fresh and/or
15 carbonised rice husk it would be apparent to a person skilled in the art that other cereal husks such as sorghum husk may be suitable.

Throughout the description and claims of the specification where reference is made to the dosage of fresh and/or carbonised rice husk, this dosage is expressed in terms of "phr" (parts per hundred) based on the weight of the composite material into which the rice husk is being introduced.

- 5 Throughout the description and claims of the specification the word "comprise" and variations of the word, such as "comprising" and "comprises", is not intended to exclude other additives, components, integers or steps.

CLAIMS:

1. A filler for use in composite materials wherein said filler comprises a vegetative-based material and wherein said vegetative-based material can be
5 fresh or carbonised.
2. A filler according to claim 1 wherein said vegetative-based material comprises cereal husk.
- 10 3. A filler according to claim 2 wherein said cereal husk comprises carbonised cereal husk.
4. A filler according to claim 3 wherein said carbonised cereal husk comprises carbonised rice husk.
15
5. A filler according to claim 4 wherein said carbonised rice husk is the product of burning fresh rice husk at about 800°C.
6. A filler according to claim 5 wherein said carbonised rice husk is the
20 product of burning fresh rice husk at about 800°C for about 4 seconds.
7. A filler according to claim 6 wherein the carbonised rice husk is the product of burning fresh rice husk at 803 to 804°C for 3 to 4 seconds.
- 25 8. A process for the production of a carbonised vegetative-based filler wherein said process comprises burning a fresh vegetative-based material at about 800°C for about 4 seconds.
9. A process according to claim 8 wherein the fresh vegetative material is
30 ground to a particle size of from 100 mesh to 400 mesh.

10. A process according to claim 8 or claim 9 wherein said fresh vegetative-based material is burned at about 803 to 804°C for 3 to 4 seconds.

11. A process according to any one of claims 8 to 10 wherein said process
5 utilises fresh rice husk as the vegetative material.

12. A process according to claim 11 wherein the fresh cereal husk comprises fresh rice husk.

10 13. A filler according to any one of claims 1 to 7 when produced by a process according to any one of claims 8 to 12.

14. A method for improving the anti-static, flame retardant, accelerator, plasticiser and/or blowing characteristics of a composite material wherein said
15 method comprises blending into the composite material with a carbonised vegetative-based filler according to any one of claims 1 to 7 and wherein said blending is substantially completed prior to incorporation of any additives, if any.

15. A method according to claim 14 wherein the carbonised vegetative filler
20 has a particle size of from 100 mesh to 400 mesh.

16. A method according to claim 14 or claim 15 wherein the composite material is latex (NR/SR) the dosage of the carbonised vegetative filler is from 1.5 to 2.5 phr (parts per hundred).

25

17. A method according to any one of claims 14 to 16 wherein said composite material is selected from the group comprising:

- i) thermoplastic resins;
- ii) thermoset plastics;
- 30 iii) rubbers and elastomeric materials;
- iv) conductive coatings;
- v) printing inks;
- vi) bitumen; and

vii) concrete.

18. A composite material having improved anti-static, flame retardant, accelerator, plasticiser and/or blowing characteristics wherein said composite
5 material is produced by the method of any one of claims 14 to 17.

19. A method for improving the mechanical properties of bitumen, said method comprising blending fresh rice husk into said bitumen.

10 20. A method according to claim 19 wherein the rice husk has a particle size of from 100 to 200 mesh and the dosage of rice husk is between 40 to 60 phr.

21. A method for improving the blowing character of a thermoplastic resin this method comprising blending fresh and/or carbonised rice husk into said
15 thermoplastic resin.

22. A method according to claim 21 wherein the rice husk has a particle size of between 325 to 400 mesh and the dosage of the rice husk is between 1.5 to 2.5
20 phr.

23. A method for improving the mechanical properties of thermoplastic resin including compression strength, said method comprising blending rice husk into said thermoplastic resin.

24. A method according to claim 23 wherein the rice husk as a particle size of between 325 to 400 mesh and the dosage of the rice husk is between 1.5 and 2.5
25 phr.

25. A method for improving the blowing character of rubber said method
30 comprising blending fresh and/or carbonised rice husk into said rubber.

26. A method according to claim 25 wherein the rice husk has a particle size of between 325 to 400 mesh and the dosage of the rice husk is between 1.5 and 27 phr.

5 27. A method for reducing the cure time of ebonite NR wherein said method comprises blending fresh rice husk into said ebonite NR.

28. A method according to claim 27 wherein the rice husk has a particle size of between 100-200 mesh and the dosage of the rice husk is between 16 to 30 phr.

10

29. A method for improving the scotch time of rubber said method comprising blending fresh and/or carbonised rice husk into said rubber.

15 30. A method according to claim 29 wherein the rice husk has a particle size of between 100 to 200 mesh and the dosage of rice husk is between 5 to 10 phr.

31. A method for improving the flame retardant ability of a thermoset resin said method comprising blending carbonised rice husk into said thermoset resin.

20 32. A method according to claim 31 wherein the carbonised rice husk has a particle size of 325 to 400 mesh and the dosage of carbonised rice husk is between 10 to 15 phr.

25 33. A method for improving the mechanical properties of thermoset resins including tensile and torque strength, said method comprising blending fresh and/or carbonised rice husk into said thermoset resin.

34. A method according to claim 33 wherein the rice husk has a particle size of between 100 to 200 mesh and the dosage of rice husk is between 10 to 15 phr.

30

35. A method for improving the antistatic properties of a thermoset resin said method comprising blending carbonised rice husk into said thermoset resin.

36. A method according to claim 34 wherein the carbonised rice husk has a particle size of between 325 to 400 mesh and the dosage of carbonised rice husk is between 10 to 15 phr.

5 37. A method for improving the antistatic properties of rubber and/or rubber-latex said method comprising blending carbonised rice husk into said rubber and/or rubber-latex.

10 38. A method according to claim 37 wherein the carbonised rice husk has a particle size of between 325 to 400 mesh and the dosage of carbonised rice husk is between 5 to 15 phr.

39. A method for improving the mechanical properties of concrete said method comprising blending fresh and/or carbonised rice husk into said concrete.

15

40. A method according to claim 39 wherein the rice husk has a particle size of between 100 to 200 mesh and the dosage of the rice husk is between 10 to 15 phr.

20 41. A filler according to claim 1 substantially as hereinbefore described with reference to any of the examples.

42. A process according to claim 8 substantially as hereinbefore described with reference to any of the examples.

25

43. A method according to claim 14 substantially as hereinbefore described with reference to any of the examples.

30 44. A composite material according to claim 18 substantially as hereinbefore described with reference to any of the examples.

PATENT COOPERATION TREATY
PCT
INTERNATIONAL PRELIMINARY EXAMINATION REPORT
(PCT Article 36 and Rule 70)

Applicant's or agent's file reference IRN606621	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416).
International Application No. PCT/AU00/00018	International Filing Date (<i>day/month/year</i>) 17 January 2000	Priority Date (<i>day/month/year</i>) 18 January 1999
International Patent Classification (IPC) or national classification and IPC Int. Cl.⁷ C08L 97/02 C08K 3/36		
Applicant CONTRACT RESEARCH & DEVELOPMENT (M) SDN. BHD. et al		

1.	This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.																								
2.	This REPORT consists of a total of 4 sheets, including this cover sheet. <input checked="" type="checkbox"/> This report is also accompanied by ANNEXES, i.e., sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT). These annexes consist of a total of 22 sheet(s).																								
3. This report contains indications relating to the following items: <table style="width: 100%; margin-top: 10px;"> <tr> <td style="width: 5%;">I</td> <td style="width: 5%;"><input checked="" type="checkbox"/></td> <td>Basis of the report</td> </tr> <tr> <td>II</td> <td><input type="checkbox"/></td> <td>Priority</td> </tr> <tr> <td>III</td> <td><input type="checkbox"/></td> <td>Non-establishment of opinion with regard to novelty, inventive step and industrial applicability</td> </tr> <tr> <td>IV</td> <td><input type="checkbox"/></td> <td>Lack of unity of invention</td> </tr> <tr> <td>V</td> <td><input checked="" type="checkbox"/></td> <td>Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement</td> </tr> <tr> <td>VI</td> <td><input type="checkbox"/></td> <td>Certain documents cited</td> </tr> <tr> <td>VII</td> <td><input type="checkbox"/></td> <td>Certain defects in the international application</td> </tr> <tr> <td>VIII</td> <td><input type="checkbox"/></td> <td>Certain observations on the international application</td> </tr> </table>		I	<input checked="" type="checkbox"/>	Basis of the report	II	<input type="checkbox"/>	Priority	III	<input type="checkbox"/>	Non-establishment of opinion with regard to novelty, inventive step and industrial applicability	IV	<input type="checkbox"/>	Lack of unity of invention	V	<input checked="" type="checkbox"/>	Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement	VI	<input type="checkbox"/>	Certain documents cited	VII	<input type="checkbox"/>	Certain defects in the international application	VIII	<input type="checkbox"/>	Certain observations on the international application
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II	<input type="checkbox"/>	Priority																							
III	<input type="checkbox"/>	Non-establishment of opinion with regard to novelty, inventive step and industrial applicability																							
IV	<input type="checkbox"/>	Lack of unity of invention																							
V	<input checked="" type="checkbox"/>	Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement																							
VI	<input type="checkbox"/>	Certain documents cited																							
VII	<input type="checkbox"/>	Certain defects in the international application																							
VIII	<input type="checkbox"/>	Certain observations on the international application																							

Date of submission of the demand 26 June 2000	Date of completion of the report 6 December 2000
Name and mailing address of the IPEA/AU AUSTRALIAN PATENT OFFICE PO BOX 200, WODEN ACT 2606, AUSTRALIA E-mail address: pct@ipaaustralia.gov.au Facsimile No. (02) 6285 3929	Authorized Officer N.L. KING Telephone No. (02) 6283 2150

I. Basis of the report

1. With regard to the elements of the international application:*
- ☐ the international application as originally filed.
- ☒ the description, pages , as originally filed,
pages **1-8 and 11-17** received on **9 October 2000** with the letter of **6 October 2000**
pages **9,10** received on **20 November 2000** with the letter of **17 November 2000**
- ☒ the claims, pages , as originally filed,
pages , as amended (together with any statement) under Article 19,
pages , filed with the demand,-
pages **18-22** received on **20 November 2000** with the letter of **17 November 2000**
- ☐ the drawings, pages , as originally filed,
pages , filed with the demand,
pages , received on with the letter of
- ☐ the sequence listing part of the description:
pages , as originally filed
pages , filed with the demand
pages , received on with the letter of
2. With regard to the language, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.
These elements were available or furnished to this Authority in the following language which is:
- ☐ the language of a translation furnished for the purposes of international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of the translation furnished for the purposes of international preliminary examination (under Rules 55.2 and/or 55.3).
3. With regard to any nucleotide and/or amino acid sequence disclosed in the international application, was on the basis of the sequence listing:
- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- / ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished
4. ☐ The amendments have resulted in the cancellation of:
- ☐ the description, pages
- ☐ the claims, Nos.
- ☐ the drawings, sheets/fig.
5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).**

* Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17).

** Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**1. Statement**

Novelty (N)	Claims 1-37	YES
	Claims	NO
Inventive step (IS)	Claims 1-37	YES
	Claims	NO
Industrial applicability (IA)	Claims 1-37	YES
	Claims	NO

2. Citations and explanations (Rule 70.7)

D1 AU 68923/94
 D2 AU 60599/94
 D3 AU 34510/95
 D4 GB 2084212
 D5 JP 4045156
 D6 JP 1249617
 D7 US 3988270
 D8 SU 956502
 D9 JP 2196012
 D10 WO 95/21550
 D11 US 4202803
 D12 JP 55139441
 D13 US 4496674
 D14 BE 1000901
 D15 US 4105459
 D16 WO 92/00251
 D17 US 3951907
 D18 GB 955049

NOVELTY(N) and INVENTIVE STEP(IS) Claims 1-11 and 24-33

Claim 3 defines a process for the production of a carbonised filler comprising the burning of fresh vegetatively-based material at about 803° to 804°C for 3 to 4 seconds while claim 1 defines a filler produced by this process.

Although D6 carbonises rice hulls at temperatures up to 800°C, there is no suggestion that carbonisation be carried out within the narrow temperature and time ranges defined in claims 1 and 3. Consequently, the above claims are novel and involve an inventive step.

NOVELTY(N) and INVENTIVE STEP(IS) Claims 12-16

Claim 12 defines a method for improving the mechanical properties of bitumen comprising blending rice husk and tyre crumb into bitumen.

Although D18 describes compositions of bitumen and rice hulls there is no teaching to blend tyre crumb into such a composition. Consequently, the above claims are novel and involve an inventive step.

Supplemental Box

(To be used when the space in any of the preceding boxes is not sufficient)

Continuation of Box V**NOVELTY(N) and INVENTIVE STEP(IS) Claims 17-23**

These claims define methods for improving various properties of thermoplastic resin, rubber and ebonite by the addition of rice husk. However, none of the above citations teach any improvement in blowing character or scotch time of rubber as defined in claims 19 and 22. Furthermore, none of the above citations use rice husk of the mesh size and dosage as defined in claims 17, 18, 20, 21 and 23. Consequently, the above claims are novel and involve an inventive step.

USE OF VEGETATIVE MATERIAL AS A FILLER
IN COMPOSITE MATERIALS

TECHNICAL FIELD

5 This invention relates to the use of a filler derived from cereal husk, more particularly rice husk, in composite materials to enhance the flame retardant, antistatic, accelerator, plasticiser and blowing characteristics in various composite materials. The invention has particular but not exclusive application to the following families of composites:-

- 10 1. Thermoplastic Resins
2. Thermoset Plastics
3. Rubbers and Elastomeric Materials
4. Conductive Coatings and Printing Inks
5. Bitumen
15 6. Concrete

BACKGROUND ART

Composite materials are well known. Fillers are usually added to composite materials, including composite polymers, to save cost or to enhance a particular mechanical property or other characteristic of the materials. The usage of fillers is usually accompanied by coupling agents that enhance the polymer-filler and filler-filler interaction so that the expected properties are realised.

The present invention is concerned with fillers which enhance the anti-static, flame retardant, accelerator, plasticiser, blowing characteristic and/or other physical or mechanical properties of composite materials and has particular application for use in composite polymers. Such have wide application in the aeronautical, mining, computer, road building, textile, foot ware, rubber and polyurethane industries among others. For example, it is often desirable to prevent the build up of static charges which can cause sparks (and hence explosions or electrical damage) or production problems, eg. collection of dust and poor feeding of materials through machinery. More highly conductive composite polymers can also be used for Electro Magnetic Interference shielding, for example.

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Carbon black fillers, aluminium flakes and fibres, stainless steel fibres and chopped carbon fibres have all been used as fillers for the purpose of rendering composite plastic conductive. Likewise other chemicals such as Halogen compounds or triethyl phosphate have been used to achieve the flame retardant property.

A number of theories have been proposed to explain how discreet particle fillers impart conductivity and flame retardant properties in composite plastics.

In order for current to flow in a conductive polymer compound, electrons must travel along the filler as the plastic itself is an excellent insulator. To achieve this flow the discreet particles of the filler must be in contact or separate by a minimum distance which is probably less than 100 Angstroms. There are three properties of the filler particles which will effect the average inter-particle distance for a given filler loading in a polymer system. These are particle size, shape (structure), and porosity. Smaller size, irregular shape and high porosity all result in smaller inter-particle distances and hence higher conductivity. A fourth property of the particle which is relevant to conductivity and flame retardant properties in the composite plastics is surface chemistry, that is the presence of oxygen on the surface. The presence of appreciable quantities of oxygen on the surface (called volatile content) acts as insulation and hence reduces conductivity.

The known conductive fillers such as carbon black, aluminium, stainless steel and carbon fibres are expensive and furthermore some of these materials have other processing difficulties, eg. aluminium fibres and stainless steel fibres settle in liquid environments due to their high density. Further problems with known conductive fillers are that they often compromise other properties of composite plastics such as flame retardance and strength.

Static electrification of articles can lead to a number of undesirable effects including:

- Attraction of dust particles.
- Attraction between surfaces, e.g. plastic films and textile yarns.
- Risk of fire or explosion caused by sparking near inflammable liquids, gases, and explosive dusts, e.g. coal dust and flour.
- Risk of shock to persons handling equipment.

- 3 -

The accumulation of electrostatic charges can be prevented by using materials of low resistance. The resistivity of natural rubber can be lowered by compounding with suitable ingredients. Alternatively, as static electrification is a surface phenomenon, the product can be covered with a conducting surface layer.

Low resistance rubber is required for a wide range of applications, such as rollers for textile machinery, conveyor belting, fuel hoses, flooring, footwear, antistatic gloves (electronic industry), cables, equipment used in hospital operating theatres, and aircraft components.

The terms "antistatic" and "conductive" are restricted here to rubber products rather than the rubber itself because the electrical resistance of the product depends not only on the resistivity of the rubber but also on the shape and most probable positions of charge generation and discharge.

Natural rubber is normally considered to be an electrically insulating material but it can be an electrically insulating material but it can be compounded to give electrical resistivity lying anywhere between 1 ohm/cm. and 10^{15} ohm/cm. The most common means of reducing resistance is to add a suitable carbon black (super conductive furnace). Resistance falls with a decrease in particle size, increase in black "structure" and increase in concentration. For light coloured products certain grades of aluminium silicate may be used as antistatic fillers although these are usually less effective in reducing resistance than the super conductive furnace. There are also other proprietary antistatic agents that are available, such as ethylene oxide, but still these agents are less effective than the super conductive furnace.

The applicant has found that carbonised rice husk is particularly suited for use as a filler in plastics as it has been found to enhance the conductivity and flame retardant properties of the composite plastics.

Honeycomb structure of a matrix is supposed to be one of the strongest structures that have been determined by Structural Engineers. The strength comes about from the full depth hexagons and half-depth trapezoids. This type of structures is presently used as designs for building bridge decks.

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The rice husk has a similar type of honeycomb design, which results in not only providing strength to the matrix, but also has sound and thermal insulation properties. The Sound insulation property is provided by the micro-cellular structures formed by the honeycomb structure in the brown rice husk. Thus the sound is trapped within the micro-cellular structure. This property is inherent to the brown (fresh) rice husk. The Thermal insulation property is provided by the honeycomb structure, which is strengthened by the silica and fibre which predominately present in brown rice husk and lesser in the carbonised (depend to the rate of carbonising) rice husk.

The presence of appreciable quantities of oxygen on the surface of carbonised rice husk acts as insulation for each aggregate, thereby reducing the conductivity and also reducing the flammability. The presence of nitrogen and oxygen in the fresh husk not only enables the blowing effect but also nitrogen being inert reduces the flame spread. The volume of gas (nitrogen/oxygen) evolution per gram of fresh rice husk is 240 ml/g. The husk's decomposition temperature is at about 280°C and curing temperature of rubber and ethyl vinyl acetate is between 130°C-180°C, thus when urea (dinitroso pentamethylene tetramine) is milled along the decomposition temperature is reduced within the curing temperatures. The presence of silica in the rice husk provides better mechanical strength.

Typical chemical and physical properties of fresh and carbonised rice husk are detailed as follows:

- consists of 20-23% of paddy
- husk burning: 20% ash by weight
90-95% is silica (amorphous and crystalline)
- physical characteristics: bulk density 96.12-112.14 kg/m³
- pH 7.14 (husk ash)
- moisture content 5.6-7.2%, dry basis
- ash 22.2%

30

AMENDED SHEET
IPE/AU

- 5 -

Chemical Composition

	Moisture Content	: 5.6-7.2%, dry basis
	Ash:	: 22.2%
	Protein	: 2.4%
5	Crude fat	: 0.7%
	Carbohydrate	: 32.0%

		Fresh RH	Carbonised RH
	Al ₂ O ₃	0.025%	0.023%
10	CaO	0.36%	0.12%
	NaO	0.034%	0.018%
	SiO ₂	96.2%	53.88%
	Fe ₂ O ₃	0.041%	0.022%
	MgO	0.16%	0.078%
15	K ₂ O	0.69%	0.95%
	P ₂ O ₅	0.57%	0.27%

It is an object according to one aspect of the present invention to provide an alternative filler which will enhance the antistatic, flame retardant, accelerator, plasticiser blowing and/or other physical or mechanical in composite materials. The filler is desirably cheap, environmentally friendly and replenishable and it does not compromise other characteristics of the composite material.

SUMMARY OF INVENTION

The present invention in one aspect resides in the use of carbonised vegetative-based filler to provide improved antistatic, flame retardant, accelerator, plasticiser, blowing and/or other physical or mechanical characteristics in composite materials.

Preferably, the carbonised vegetative-based filler is carbonised rice husk.

Preferably, the carbonised rice husk is burnt at about 800°C for about 4 seconds. Most preferably, the carbonised rice husk is burnt at 803-804°C for 3-4 seconds.

In another aspect the invention resides in a composite material, more particularly a composite plastic including a vegetative-based filler when used as a conductive or flame retardant article.

Preferably, the carbonised vegetative-based filler is carbonised rice husk which has been burnt at 803-804°C for 3-4 seconds.

The present invention also exhibits the usage of fresh and carbonised rice husk as a blowing agent when used in combination with recycled (reclaimed), or virgin natural rubber or other suitable thermoplastic materials. Though other conventional blowing agents have been used with natural or synthetic rubber to achieve the similar products but so far no blowing agents have been used with recycle (reclaim) rubber to produce similar products. Furthermore the conventional blowing agents are expensive and dosages are higher as compared to the fresh rice husk. For example for the conventional blowing agent, the dosage is about 6.5-7 phr, whereas the fresh/carbonised rice husk, the dosage is between 1.5 to 3 phr. When rice husk is used at different dosages the blowing effect is different. It was also noted that the rice husk does not only work as a blowing agent, but also as a plasticizer and a filler. The properties achieved are comparable to conventional blowing agents, when using fresh or carbonised rice, has no difference to the conventional blowing agent other than the colour of the end product.

Ebonite, a hard, dark-coloured plastic-like material, is the reaction product of rubber and a large proportion of sulphur. Simple rubber / sulphur mixtures are seldom used in practice; they suffer from poor processability, require long cure times and lead to excessive shrinkage and heat evolution during cure. Accelerators, fillers, processing aids and other compounding ingredients are widely used in ebonite, as in soft rubber vulcanised rubber, to ease processing, shorten cure times and modify properties. The curing times for ebonite are generally up to ten (10) hours at 150° C, thus making ebonite products expensive. Ebonite can be made from synthetic, such as BR, NBR, SBR and Nitrile rubber and as well as from Natural rubber. High strength, low thermal conductivity, chemical resistance and insulating properties of natural rubber make it a popular choice. Although it has been superseded in many applications by synthetic thermoplastic and thermosets, it is still used for outstanding chemical resistance and electrical properties coupled with high mechanical strength and ease of machining.

The present invention exhibits the usage of fresh and carbonised rice as an accelerator when used in combination with recycled (devulcanised) or virgin natural rubber, and at the same time making ebonite a conductive product when

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carbonised rice husk is used. Though other conventional accelerators have been used with natural or synthetic rubber (virgin or recycled) to achieve the similar products but so far no accelerators like the rice husk material have been used with recycled (devulcanised) rubber to produce similar products. Further more
5 the conventional accelerators and conductive carbon black are expensive and difficult to blend and process. When rice husk is used singularly at different dosages the activation effect is different to meet industrial requirements. Generally for ebonite production the sulphur content should be in the range of 25-40 phr, but whereas when fresh rice husk between 25-30 phr is used the sulphur
10 content could be reduced to 20-25 phr. Accelerators are less effective in ebonite than in soft rubber and large quantities are generally required. Basic accelerators such as guanidines and aldehyde-amines are preferred. Inorganic activators such as magnesium oxide, magnesium carbonate and lime are also effective when used with organic accelerators to reduce cure time without the risk of over
15 heating.

Common inorganic fillers used in ebonite are china clay, talc, silica, whiting and magnesium oxide. These also reduce shrinkage and heat evolution but loaded ebonite generally have weaker mechanical properties than unloaded ones. Carbon black does not reinforce ebonite and is normally only added for
20 pigmentation, although conductive carbon black are sometimes used to prepare electrically conducting ebonite.

BEST MODE

Following is an example of the invention, in this example the filler is
25 carbonised rice husk (CRH) which has been burnt at 803-804°C for 3-4 seconds. After this the CRH is obtained.

The manner in which the rice husk is burnt is believed to be important in achieving the desired surface area, surface structure and porosity necessary for conductivity and flame retardant and blowing properties in the composite plastics
30 to be achieved. At this stage the range of temperature and the duration of the time of burning which achieves the desired result has not been fully explored, however it is predicted that the temperature range will be from about 100-950°C and the time range will be from about 2-30 seconds, although these ranges may

be much narrower. The importance of controlled burning in a prescribed time results in obtaining better surface area and porosity which in turn offers ideal properties emitting anti-static, flame retardant and enhancing physical properties of the material. In the absence of controlled burning, the surface area, surface structure and porosity would be distorted. While the present exemplification involves use of carbonised rice husk it is possible that the desired results may be achieved by use of other carbonised vegetative-based fillers.

EXAMPLE 1

A thermoset application called pulforming was used to manufacture fibre reinforced bolts. Fibre glass tows (36 tow of 8000 tex) are pulled into a resin bath that contains:

1. Polyester and Vinyl Ester combination, ie. 60% Vinyl Ester (Derakane 411 - Dow Chemical) and 40% Polyester (Everpol 3260 AR - P.T. Arinde).
2. Zinc Stearate (mould releasing agent) - 1.18% of the resin weight
BYK 980 (improves wetting and dispersing of fillers in glass fibre reinforcement compounds) - 1.5% of the filler weight.
3. BYK 515 (air releasing agent) - 0.5% of the total weight of the resin mixture.
4. BYK 996 (wetting and dispersing additive for mineral fillers in hot curing, glass fibre reinforced UP-resin systems) - 2% of the resin weight.
5. Fillers ($\text{Ca}_2(\text{CO})_3$ & Carbonised Rice Husk (mesh size 325) @ 55% & 12% of the resin weight).
6. Aluminium Trihydrate (2.4% of the resin weight).
7. Catalyst TBPH (Tertiary Butyl Peroxy-2-Ethyl Hexanoate)
- 2.12% of the resin weight.
TBPB (Tertiary Butyl Perbenzoate)
- 0.53% of the resin weight.

The wet fibre glass tow is pulled into the mould and compressed at a pressure of 800psi (5600kPa) for 3.8 minutes at 130°C. Then the bolt is pulled out of the mould and left to cure.

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The following day tests were carried out on the bolt with the following results:

Tensile strength at the thread - 50kN

Torque - 45ft/lb

5 Bond strength - BS 1610:Part 1, Grade 1.0

Fire rating - BS 5865:1980 - Persistence flame shall be less than 10 seconds

Electrical conductivity - less than 10 to the power of 9 Ohms.

10 EXAMPLE 2

All chemicals used are taken by percentage of weight of rubber. The rubber and the chemicals are mixed in a Banbury, for 5 minutes. Recycled rubber (reclaim) (220g) is first milled with zinc oxide (4.5%) - accelerator, which is followed with stearic acid (1.8%) - activator, Mercadibenzothiazole disulphide (MBTS)(0.5%), Tetramethylthiuram disulphide (TMTD)(0.2%)- accelerator, fresh
15 rice husk (27%)-blowing agent and filler and sulphur (2.7%)- vulcanisate. Then the mixed compound is milled for five (5) minutes to form a sheet that is ready for curing. Then a piece of the sheet weighing about 32g is placed in a mould that it is to be cured for two (2) minutes in a oven at 150°C temperature. The
20 conventional curing time is six (6) minutes at the same temperature of 150°C.

The rubber and the chemicals are mixed in a Banbury, for 5 minutes. The similar approach has been done for using SBR Rubber (100g), silica (62g), Peg 1500 (2.5g), Paraffin oil (5g), Zinc oxide (2.5g), Wing stay (0.5g), Wax (1g),
25 Mercadibenzothiazole disulphide (MBTS) (1.5g), Tetramethylthiuram disulphide (TMTD) (0.2g), Stearic acid (1.5g) and Sulphur (2g). The milling was done for ten (10) minutes and later cured in the oven for six (6) minutes at 150°C.

This exercise was repeated by using fifty (50) percent of the virgin material compound and fifty (50) percent recycled (reclaimed) material compound, and
30 cured in the oven at 150°C for two (2) minutes.

With the level, of rice husk dosage, the blowing effect can be controlled to suit the industry's requirement.

**AMENDED SHEET
IPEA/AU**

Machine Operating Conditions

Virgin Rubber:

5

PROPERTIES	SMR-10	TYRE DUST	TYRE DUST WITH BROWN RICE HUSK	EXAMPLE 1 WITH BROWN RICE HUSK	EXAMPLE 1 WITHOUT BROWN RICE HUSK
Mooney Viscosity MLI + 3, 100°C	60	36.6	31.8	24	30
Monsanto Rheometer, 150°C					
Scorch time	1.4	3.8	1.2	2	2.5
Cure time	7.4	3.5	3.25	4	4

By using rice husk the Mooney viscosity was lower than the conventional filler, thus lowering scorch time (time taken by the rubber compound to flow into the mould) and curing time (time taken to cure rubber compound) respectively. As such this leads to a cheaper production system. Presently various fillers and blowing agents are being used in the production of soft/spongy rubber that would produce different types of cell structures for an end product, but the cost determines the market.

10

EXAMPLE 3

All chemicals used are taken by percentage of weight of rubber. The rubber and the chemicals are mixed in a Banbury, for six (6) minutes. The recycle (devulcanised) rubber is first milled with magnesium oxide (2%) - accelerator, which is followed with Diphenylguanidine (2%) - accelerator, fresh rice husk (30%) - accelerator and filler and sulphur (30%) - vulcanisate. After the milling at the Banbury for ten (10) minutes, it is then milled into a sheet. The mould was heated in the oven press to 150°C then the sheeted rubber is placed in the mould and it is cured for twelve (12) minutes. The conventional curing time is between eight to ten hours at the same temperature of 150°C.

A conventional formula for ebonite was selected to compare. The rubber and the chemicals are mixed in a Banbury, for 5 minutes. The similar mixing as above was followed, using SBR 5 Rubber (100g), ebonite dust (100g), China clay (50g), Magnesium oxide (5g), Diphenylguanidine (3g), Linseed oil (5g) and Sulphur (45g). The milling was done for ten (10) minutes and later cured in the oven for eight (8) hours at 150°C.

Mix properties; Rice Husk filled mix

20	Mooney viscosity, MLI + 3, 100°C	24
	Mooney viscosity, MLI + 3, 120°C	18.5
	Mooney scorch, t ₅ , MLI + 3, 120°C min.	5.8
	Monsanto Rheometer, 160°C	
	time to 95% cross-linking, s	110

25

By using rice husk the curing time is reduced tremendously twelve minutes as compared to eight to ten hours. The sulphur content in the rubber polymer is reduced by fifteen percent.

The results are based on cure time, the formulation with rice husk cures faster than the formulation without rice husk, i.e. twelve minutes for with brown rice husk and about eight hour without rice husk.

EXAMPLE 4

All chemicals used are taken by percentage of weight of natural rubber (NR). The natural rubber and the chemicals are mixed in an open mill or kinder, for six (6) minutes. Natural rubber is first milled with stearic acid (1%) and zinc oxide (5%) activator, which is followed with rice husk (blowing agent) (2.5-3.5%), calcium carbonate - (40%), promoter - urea based (2.5-3.5%), silica (10%), accelerator dibenzthiazyl disulphide (MBTS) (0.05%) and catalyst sulphur (1.5%). After the milling at the open mill or kinder for ten (10) minutes, it is then milled into a sheet. The mould was heated in the oven press to 160°C then the sheeted natural rubber is placed in the mould and it is cured for twenty-two (22) minutes.

The temperature for curing could be from 145°-160°C and the cure time may differ according to the mould size.

15 Cured properties; rice husk filled blowed mix - micro-cellular cells.

1.	Hardness	Askar C	35
2.	Shrinkage	%	5
3.	Specific Gravity	g/cc	0.3-0.35

20 By using rice husk as a blowing agent the catalyst percentage could be reduced and as well as the percentage of blowing agent used.

EXAMPLE 5

THERMOPLASTIC (EVA)

25 All chemicals used are taken by percentage of weight of thermoplastic - Ethyl Vinyl Acetate (EVA). The EVA and the chemicals are mixed in an Open Mill or Knider, for six (6) minutes. Ethyl Vinyl Acetate (EVA) is first milled with Stearic Acid (1%) and zinc oxide (5%) accelerator, which is followed with Rice Husk (Blowing agent) (2.5%). Magnesium carbonate - (10%), Promoter - urea based (5%) and catalyst Diacyl Peroxide (1%). After the milling at the Open mill or Knider for ten (10) minutes, it is then milled into a sheet. The mould was heated in the oven press to 160°C then the sheeted EVA is placed in the mould and it is cured for twenty-two (22) minutes.

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The temperature for curing could be from 145°-160°C and the cure time may differ according to the mould size.

Cured properties: Rice Husk filled blowed mix - micro-cellular cells

	1.	HARDNESS	Askar C	29-35
5	2.	SHRINKAGE	%	2
	3.	SPECIFIC GRAVITY	g/cc	0.200
	4.	COMPRESSION SET	%	80

By using rice husk as a blowing agent the catalyst percentage could be reduced and as well as the percentage of blowing agent used.

EXAMPLE 6

The rice husk is mixed by weight with tyre crumbs(35-40 mesh) and an effluent from the palm oil mill called Scavenger (which have a fatty acid content (C₈-C₁₈). From literature it has been reported that by using tyre crumb with the binder (bitumen) there is an improve of properties for the asphalt mixture. This invention not only uses tyre crumb along with rice husk and an oil palm effluent to further improve the properties. As well as the formulation address the recyclability of all agro waste by-products to be used in the road surfacing industries. The formulation of the rice husk mixture as follows:

Rice husk	50%
Tyre crumb	45%
Scavenger	5%

In this particular example the usage of rice husk mixture is divided into two categories:

- A. RICE HUSK MIXTURE USED IN MODIFIED BINDER
- B. RICE HUSK MIXTURE USED IN AS FILLER

30

- 14 -

- 5 A. The rice husk mixture is added to the bitumen first in compliance to the
SOCIETY OF HIGHWAY PROCEDURE (SHRP) to manufacture modified
bitumen. The bitumen is first heated to about 160°C, then the rice husk
mixture twenty percent 20% by weight of bitumen is mixed with the heated
bitumen for about one hour. As a result of this reaction a modified bitumen
is made. From here 5-7% by weight of this modified bitumen is added to
the aggregate. The aggregate is first heated to about 200°C and the
modified bitumen is mixed for three to four minutes. The modified bitumen
with rice husk mixture complies to all requirement of the SHRP.
- 10 B. The rice husk mixture is added as a filler to the aggregate, by four (4%) by
weight to the aggregate weight. The aggregate is first heated to 200°C,
and is allowed to cool to about 160°C, then the rice husk mixture is added
and mixed and lastly the bitumen 5-6% by weight of aggregate is added
and mixed for 3-4 minutes. This blending with rice husk mixture complies
15 to all requirement of the Marshall Properties.

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TABLE 1

PROPERTIES OF RICE HUSK MODIFIED BINDER - SHRP

SHRP	80/100	RICE HUSK MIXTURE
FLASH POINT TEMP. °C	230	240
SOFTENING POINT, °C	44-50	55-70
PENETRATION @ °C 25, dmm	80-100	60-100
BROOKFIELD VISCOSITY @ 135°C, MPaS	<500	>1500
DYNAMIC SHEAR RHEOMETER		
PG 70		
ORIGINAL		
G* (Pa)	<1000	>1200
$\delta(^{\circ})$	>80	>80
G*/Sin δ	<1000	>1200
AFTER RTFOT		
G* (Pa)	<1000	>3800
$\delta(^{\circ})$	>80	>70
G*/Sin δ	<1000	>3800
AFTER PAV		
G* (Pa)	<1000	>230
$\delta(^{\circ})$	>80	>50
G*/Sin δ	<1000	>260
PG 76		
ORIGINAL		
G* (Pa)	<1000	>1800
$\delta(^{\circ})$	>80	>70
G*/Sin δ	<1000	>1800
AFTER RTFOT		
G* (Pa)	<1000	>2400
$\delta(^{\circ})$	>80	>70
G*/Sin δ	<1000	>2600
AFTER PAV		
G* (Pa)	<1000	>230
$\delta(^{\circ})$	>80	>50
G*/Sin δ	<1000	>280

TABLE 2

MIXED PROPERTIES OF RICE HUSK MODIFIED BINDER

5

PROPERTIES	80/100	RICE HUSK MIXTURE
MARSHALL STABILITY (kN)	5-10	>13
FLOW (mm)	2-4	2-4
QUOTIENT (kN/mm)	1-3.5	3-4
RESILIENT MODULUS @ 25°	>2000	>2800

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TABLE 3

MIXED PROPERTIES OF RICE HUSK MIXTURE AS FILLER

PROPERTIES	80/100	RICE HUSK MIXTURE
MARSHALL STABILITY (kN)	6-10	>12
FLOW (mm)	2-4	2-4
QUOTIENT (kN/mm)	1-3.5	3-4
RESILIENT MODULUS @ 25°	>2000	>2800

TABLE 4

PREFERRED PARTICLE SIZE AND DOSAGE OF FRESH AND/OR CARBONISED RICE HUSK FOR PARTICULAR COMPOSITE MATERIALS

COMPOSITE MATERIAL	FRESH/DOSAGE	CARBONISED/DOSAGE
BITUMEN (MECHANICAL PROPERTY)	100 MESH - 40-60 phr	-
THERMOPLASTIC (EVA) BLOWING CHARACTER	325-400 MESH - 1.5-2.5 phr	325-400 MESH - 1.5-2.6 phr
THERMOPLASTIC (EVA) MECHANICAL PROPERTY	325-400 MESH - 1.5-5 phr	325-400 MESH - 1.5-2.5 phr
RUBBER (N.R./S.R.) BLOWING CHARACTER	325-400 MESH - 1.5-27 phr	325-400 MESH - 1.5-27 phr
EBONITE N.R. (REDUCE CURE TIME)	100-200 MESH 18-30 phr	-
RUBBER (N.R./S.R.) SCOTCH TIME	100-200 MESH 5-10 phr	100-200 MESH 5-10 phr
THERMOSET RESIN (FLAME PROPERTY)	-	325 MESH 10-15 phr
THERMOSET RESIN (MECHANICAL PROPERTY)	100-200 MESH 10-15 phr	100-200 MESH 10-15 phr
THERMOSET RESIN (ANTISTATIC)	-	325 MESH 10-15 phr
RUBBER-LATEX (N.R./S.R.) ANTISTATIC	-	325-400 MESH 5-15 phr
RUBBER (N.R./S.R.) ANTISTATIC	-	325-400 MESH 5-15 phr
CONCRETE (MECHANICAL PROPERTY)	100-200 MESH 10-15 phr	100-200 MESH 10-15 phr

N.R. - NATURAL RUBBER
S.R. - SYNTHETIC RUBBER

10 It will of course be realised that whilst the above has been given by way of illustrative examples of this invention, all such and other modifications and variations hereto, as would be apparent to persons skilled in the art, are deemed to fall within the broad scope and ambit of this invention as herein set forth. For instance, while the preceding examples relate to the use of fresh and/or
15 carbonised rice husk it would be apparent to a person skilled in the art that other cereal husks such as sorghum husk may be suitable.

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Throughout the description and claims of the specification where reference is made to the dosage of fresh and/or carbonised rice husk, this dosage is expressed in terms of "phr" (parts per hundred) based on the weight of the composite material into which the rice husk is being introduced.

- 5 Throughout the description and claims of the specification the word "comprise" and variations of the word, such as "comprising" and "comprises", is not intended to exclude other additives, components, integers or steps.

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THE CLAIMS DEFINING THIS INVENTION ARE AS FOLLOWS:

1. A filler for use in composite materials wherein said filler comprises
5 carbonized vegetative-based material wherein said carbonized vegetative-
based material is the product of burning fresh vegetative-based material at 803°
to 804°C for 3 to 4 seconds.
2. A filler according to claim 1 wherein the carbonized vegetative-based
10 material is carbonized rice husk.
3. A process for the production of a carbonised vegetative-based filler
wherein said process comprises burning a fresh vegetative-based material at
about 803° to 804°C for 3 to 4 seconds.
15
4. A process according to claim 3 wherein the fresh vegetative material is
ground to a particle size of from 100 mesh to 400 mesh.
5. A process according to claim 3 or claim 4 wherein said process utilises
20 fresh rice husk as the vegetative material.
6. A filler according to any one of claims 1 or 2 when produced by a
process according to claim 4.
- 25 7. A method for improving the anti-static, flame retardant, accelerator,
plasticiser and/or blowing characteristics of a composite material wherein said
method comprises blending into the composite material with a carbonised
vegetative-based filler according to claim 1 or claim 2 and wherein said blending
is substantially completed prior to incorporation of any additives, if any.
30
8. A method according to claim 7 wherein the carbonised vegetative filler
has a particle size of from 100 mesh to 400 mesh.

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9. A method according to claim 7 or claim 8 wherein the composite material is latex (NR/SR) the dosage of the carbonised vegetative filler is from 1.5 to 2.5 phr (parts per hundred).

5

10. A method according to any one of claims 7 to 9 wherein said composite material is selected from the group comprising:

- i) thermoplastic resins;
- 10 ii) thermoset plastics;
- iii) rubbers and elastomeric materials;
- iv) conductive coatings;
- v) printing inks;
- vi) bitumen; and
- 15 vii) concrete.

11. A composite material having improved anti-static, flame retardant, accelerator, plasticiser and/or blowing characteristics wherein said composite material is produced by the method of any one of claims 7 to 10

20

12. A method for improving the mechanical properties of bitumen, said method comprising blending fresh and/or carbonised rice husk together with tyre crumb into said bitumen.

25 13. A method according to claim 12 wherein the rice husk has a particle size of from 100 to 200 mesh and the dosage of rice husk is between 40 to 60 phr.

14. A method according to claim 12 or claim 13 wherein the rice husk and tyre crumb is mixed in composition with a palm oil effluent prior to it being
30 added to the bitumen.

15. A method according to claim 14 wherein the composition comprises about 50% tyre crumb, about 45% rice husk and about 5% palm oil effluent.

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16. A method according to claim 14 or claim 15 wherein the composition is added in a dosage amount of about 20% by weight of the said bitumen.
- 5 17. A method for improving the blowing character of a thermoplastic resin this method comprising blending fresh and/or carbonised rice husk into said thermoplastic resin. wherein the rice husk has a particle size of between 325 to 400 mesh and the dosage of the rice husk is between 1.5 to 2.5 phr.
- 10 18. A method for improving the mechanical properties of thermoplastic resin including compression strength, said method comprising blending rice husk into said thermoplastic resin wherein said rice husk has a particle size of between 325 to 400 mesh and the dosage of the rice husk is between 1.5 and 2.5 phr.
- 15 19. A method for improving the blowing character of rubber said method comprising blending fresh and/or carbonised rice husk into said rubber.
20. A method according to claim 19 wherein the rice husk has a particle size of between 325 to 400 mesh and the dosage of the rice husk is between 1.5
20 and 27 phr.
21. A method for reducing the cure time of ebonite NR wherein said method comprises blending fresh rice husk into said ebonite NR. wherein the rice husk has a particle size of between 100 to 200 mesh and the dosage of the rice husk
25 is between 16 to 30 phr.
22. A method for improving the scotch time of rubber said method comprising blending fresh and/or carbonised rice husk into said rubber.
- 30 23. A method according to claim 22 wherein the rice husk has a particle size of between 100 to 200 mesh and the dosage of rice husk is between 5 to 10 phr.

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24. A method for improving the flame retardant ability of a thermoset resin said method comprising blending carbonised rice husk according to claim 2 into said thermoset resin.

5

25. A method according to claim 24 wherein the carbonised rice husk has a particle size of 325 to 400 mesh and the dosage of carbonised rice husk is between 10 to 15 phr.

10 26. A method for improving the mechanical properties of thermoset resins including tensile and torque strength, said method comprising blending carbonised rice husk according to claim 2 into said thermoset resin.

15 27. A method according to claim 26 wherein the rice husk has a particle size of between 100 to 200 mesh and the dosage of rice husk is between 10 to 15 phr.

20 28. A method for improving the anti-static properties of a thermoset resin said method comprising blending carbonised rice husk according to claim 2 into said thermoset resin.

25 29. A method according to claim 27 wherein the carbonised rice husk has a particle size of between 325 to 400 mesh and the dosage of carbonised rice husk is between 10 to 15 phr.

30. A method for improving the anti-static properties of rubber and/or rubber latex said method comprising blending carbonised rice husk according to claim 2 into said rubber and/or rubber-latex.

30 31. A method according to claim 30 wherein the carbonised rice husk has a particle size of between 325 to 400 mesh and the dosage of carbonised rice husk is between 5 to 15 phr.

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32. A method for improving the mechanical properties of concrete said method comprising blending carbonised rice husk according to claim 2 into said concrete.

5

33. A method according to claim 32 wherein the rice husk has a particle size of between 100 to 200 mesh and the dosage of the rice husk is between 10 to 15 phr.

10 34. A filler according to claim 1 substantially as hereinbefore described with reference to any of the examples.

35. A process according to claim 3 substantially as hereinbefore described with reference to any of the examples.

15

36. A method according to claim 7 substantially as hereinbefore described with reference to any of the examples.

20 37. A composite material according to claim 11 substantially as hereinbefore described with reference to any of the examples.

DATED: 17 November, 2000

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And Mr. D J .Tadgell

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ART 34 AMBT

PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

REC'D 19 DEC 2000

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15

Applicant's or agent's file reference IRN606621	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416).
International Application No. PCT/AU00/00018	International Filing Date (<i>day/month/year</i>) 17 January 2000	Priority Date (<i>day/month/year</i>) 18 January 1999
International Patent Classification (IPC) or national classification and IPC Int. Cl. ⁷ C08L 97/02 C08K 3/36		
Applicant CONTRACT RESEARCH & DEVELOPMENT (M) SDN. BHD. et al		

1.	This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.	
2.	This REPORT consists of a total of 4 sheets, including this cover sheet. <input checked="" type="checkbox"/> This report is also accompanied by ANNEXES, i.e., sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT). These annexes consist of a total of 22 sheet(s).	
3.	This report contains indications relating to the following items: I <input checked="" type="checkbox"/> Basis of the report II <input type="checkbox"/> Priority III <input type="checkbox"/> Non-establishment of opinion with regard to novelty, inventive step and industrial applicability IV <input type="checkbox"/> Lack of unity of invention V <input checked="" type="checkbox"/> Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement VI <input type="checkbox"/> Certain documents cited VII <input type="checkbox"/> Certain defects in the international application VIII <input type="checkbox"/> Certain observations on the international application	

Date of submission of the demand 26 June 2000	Date of completion of the report 6 December 2000
Name and mailing address of the IPEA/AU AUSTRALIAN PATENT OFFICE PO BOX 200, WODEN ACT 2606, AUSTRALIA E-mail address: pct@ipaaustralia.gov.au Facsimile No. (02) 6285 3929	Authorized Officer N.L. KING Telephone No. (02) 6283 2150

I. Basis of the report1. With regard to the **elements** of the international application:*

- ☐ the international application as originally filed.
- ☒ the description, pages , as originally filed,
pages **1-8 and 11-17 received on 9 October 2000 with the letter of 6 October 2000**
pages **9,10 received on 20 November 2000 with the letter of 17 November 2000**
- ☒ the claims, pages , as originally filed,
pages , as amended (together with any statement) under Article 19,
pages , filed with the demand,-
pages **18-22 received on 20 November 2000 with the letter of 17 November 2000**
- ☐ the drawings, pages , as originally filed,
pages , filed with the demand,
pages , received on with the letter of
- ☐ the sequence listing part of the description:
pages , as originally filed
pages , filed with the demand
pages , received on with the letter of

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language which is:

- ☐ the language of a translation furnished for the purposes of international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of the translation furnished for the purposes of international preliminary examination (under Rules 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, was on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

4. ☐ The amendments have resulted in the cancellation of:

- ☐ the description, pages
- ☐ the claims, Nos.
- ☐ the drawings, sheets/fig.

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).**

* Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17).

** Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**1. Statement**

Novelty (N)	Claims 1-37	YES
	Claims	NO
Inventive step (IS)	Claims 1-37	YES
	Claims	NO
Industrial applicability (IA)	Claims 1-37	YES
	Claims	NO

2. Citations and explanations (Rule 70.7)

D1 AU 68923/94
 D2 AU 60599/94
 D3 AU 34510/95
 D4 GB 2084212
 D5 JP 4045156
 D6 JP 1249617
 D7 US 3988270
 D8 SU 956502
 D9 JP 2196012
 D10 WO 95/21550
 D11 US 4202803
 D12 JP 55139441
 D13 US 4496674
 D14 BE 1000901
 D15 US 4105459
 D16 WO 92/00251
 D17 US 3951907
 D18 GB 955049

NOVELTY(N) and INVENTIVE STEP(IS) Claims 1-11 and 24-33

Claim 3 defines a process for the production of a carbonised filler comprising the burning of fresh vegetatively-based material at about 803° to 804°C for 3 to 4 seconds while claim 1 defines a filler produced by this process.

Although D6 carbonises rice hulls at temperatures up to 800°C, there is no suggestion that carbonisation be carried out within the narrow temperature and time ranges defined in claims 1 and 3. Consequently, the above claims are novel and involve an inventive step.

NOVELTY(N) and INVENTIVE STEP(IS) Claims 12-16

Claim 12 defines a method for improving the mechanical properties of bitumen comprising blending rice husk and tyre crumb into bitumen.

Although D18 describes compositions of bitumen and rice hulls there is no teaching to blend tyre crumb into such a composition. Consequently, the above claims are novel and involve an inventive step.

Supplemental Box

(To be used when the space in any of the preceding boxes is not sufficient)

Continuation of Box V**NOVELTY(N) and INVENTIVE STEP(IS) Claims 17-23**

These claims define methods for improving various properties of thermoplastic resin, rubber and ebonite by the addition of rice husk. However, none of the above citations teach any improvement in blowing character or scotch time of rubber as defined in claims 19 and 22. Furthermore, none of the above citations use rice husk of the mesh size and dosage as defined in claims 17, 18, 20,21 and 23. Consequently, the above claims are novel and involve an inventive step.

USE OF VEGETATIVE MATERIAL AS A FILLER IN COMPOSITE MATERIALS

TECHNICAL FIELD

5 This invention relates to the use of a filler derived from cereal husk, more particularly rice husk, in composite materials to enhance the flame retardant, antistatic, accelerator, plasticiser and blowing characteristics in various composite materials. The invention has particular but not exclusive application to the following families of composites:-

- 10 1. Thermoplastic Resins
2. Thermoset Plastics
3. Rubbers and Elastomeric Materials
4. Conductive Coatings and Printing Inks
5. Bitumen
- 15 6. Concrete

BACKGROUND ART

Composite materials are well known. Fillers are usually added to composite materials, including composite polymers, to save cost or to enhance a particular mechanical property or other characteristic of the materials. The usage of fillers is usually accompanied by coupling agents that enhance the polymer-filler and filler-filler interaction so that the expected properties are realised.

The present invention is concerned with fillers which enhance the anti-static, flame retardant, accelerator, plasticiser, blowing characteristic and/or other physical or mechanical properties of composite materials and has particular application for use in composite polymers. Such have wide application in the aeronautical, mining, computer, road building, textile, foot ware, rubber and polyurethane industries among others. For example, it is often desirable to prevent the build up of static charges which can cause sparks (and hence explosions or electrical damage) or production problems, eg. collection of dust and poor feeding of materials through machinery. More highly conductive composite polymers can also be used for Electro Magnetic Interference shielding, for example.

Carbon black fillers, aluminium flakes and fibres, stainless steel fibres and chopped carbon fibres have all been used as fillers for the purpose of rendering composite plastic conductive. Likewise other chemicals such as Halogen compounds or triethyl phosphate have been used to achieve the flame retardant property.

A number of theories have been proposed to explain how discrete particle fillers impart conductivity and flame retardant properties in composite plastics.

In order for current to flow in a conductive polymer compound, electrons must travel along the filler as the plastic itself is an excellent insulator. To achieve this flow the discrete particles of the filler must be in contact or separated by a minimum distance which is probably less than 100 Angstroms. There are three properties of the filler particles which will effect the average inter-particle distance for a given filler loading in a polymer system. These are particle size, shape (structure), and porosity. Smaller size, irregular shape and high porosity all result in smaller inter-particle distances and hence higher conductivity. A fourth property of the particle which is relevant to conductivity and flame retardant properties in the composite plastics is surface chemistry, that is the presence of oxygen on the surface. The presence of appreciable quantities of oxygen on the surface (called volatile content) acts as insulation and hence reduces conductivity.

The known conductive fillers such as carbon black, aluminium, stainless steel and carbon fibres are expensive and furthermore some of these materials have other processing difficulties, eg. aluminium fibres and stainless steel fibres settle in liquid environments due to their high density. Further problems with known conductive fillers are that they often compromise other properties of composite plastics such as flame retardance and strength.

Static electrification of articles can lead to a number of undesirable effects including:

- Attraction of dust particles.
- Attraction between surfaces, e.g. plastic films and textile yarns.
- Risk of fire or explosion caused by sparking near inflammable liquids, gases, and explosive dusts, e.g. coal dust and flour.
- Risk of shock to persons handling equipment.

The accumulation of electrostatic charges can be prevented by using materials of low resistance. The resistivity of natural rubber can be lowered by compounding with suitable ingredients. Alternatively, as static electrification is a surface phenomenon, the product can be covered with a conducting surface layer.

Low resistance rubber is required for a wide range of applications, such as rollers for textile machinery, conveyor belting, fuel hoses, flooring, footwear, antistatic gloves (electronic industry), cables, equipment used in hospital operating theatres, and aircraft components.

The terms "antistatic" and "conductive" are restricted here to rubber products rather than the rubber itself because the electrical resistance of the product depends not only on the resistivity of the rubber but also on the shape and most probable positions of charge generation and discharge.

Natural rubber is normally considered to be an electrically insulating material but it can be an electrically insulating material but it can be compounded to give electrical resistivity lying anywhere between 1 ohm/cm. and 10^{15} ohm/cm. The most common means of reducing resistance is to add a suitable carbon black (super conductive furnace). Resistance falls with a decrease in particle size, increase in black "structure" and increase in concentration. For light coloured products certain grades of aluminium silicate may be used as antistatic fillers although these are usually less effective in reducing resistance than the super conductive furnace. There are also other proprietary antistatic agents that are available, such as ethylene oxide, but still these agents are less effective than the super conductive furnace.

The applicant has found that cereal husk (more preferably but not exclusively carbonised rice husk) is particularly suited for use as a filler in plastics as it has been found to enhance the conductivity and flame retardant properties of the composite plastics. The presence of appreciable quantities of oxygen on the surface of carbonised rice husk acts as insulation for each aggregate, thereby reducing the conductivity and also reducing the flammability. The presence of nitrogen and oxygen in the fresh husk not only enables the blowing effect but also nitrogen being inert reduces the flame spread. The volume of gas (nitrogen/oxygen) evolution per gram of fresh rice husk is 240 ml/g. The husk's

decomposition temperature is at about 280°C and curing temperature of rubber and ethyl vinyl acetate is between 130°C-180°C, thus when urea is milled along the decomposition temperature is reduced within the curing temperatures. The presence of silica in the rice husk provides better mechanical strength.

5 Typical chemical and physical properties of fresh and carbonised rice husk are detailed as follows:

- consists of 20-23% of paddy
- husk burning: 20% ash by weight
90-95% is silica (amorphous and crystalline)
- 10 • physical characteristics: bulk density 96.12-112.14 kg/m³
- pH 7.14 (husk ash)
- moisture content 5.6-7.2%, dry basis
- ash 22.2%

15 Chemical Composition

	Moisture Content	: 5.6-7.2%, dry basis
	Ash:	: 22.2%
	Protein	: 2.4%
	Crude fat	: 0.7%
20	Carbohydrate	: 32.0%

		Fresh RH	Carbonised RH
	Al ₂ O ₃	0.025%	0.023%
	CaO	0.36%	0.12%
25	NaO	0.034%	0.018%
	SiO ₂	96.2%	53.88%
	Fe ₂ O ₃	0.041%	0.022%
	MgO	0.16%	0.078%
	K ₂ O	0.69%	0.95%
30	P ₂ O ₅	0.57%	0.27%

It is an object according to one aspect of the present invention to provide an alternative filler which will enhance the antistatic, flame retardant, accelerator, plasticiser blowing and/or other physical or mechanical in composite materials.

35 The filler is desirably cheap, environmentally friendly and replenishable and it does not compromise other characteristics of the composite material.

SUMMARY OF INVENTION

The present invention in one aspect resides in the use of fresh and/or carbonised vegetative-based filler to provide improved antistatic, flame retardant, accelerator, plasticiser, blowing and/or other physical or mechanical characteristics in composite materials.

Preferably, the fresh vegetative-based filler is fresh/raw rice husk. Fresh rice husk is obtained after the husk is separated from the grain. The husk is ground by any mechanical process, i.e., using hammer mill or cryogenic or jet mill. As the fresh rice husk is used in various composite materials to determine various properties therefore it is imperative to determine the particle size for each usage. For example with bitumen a particle size of 100 mesh size, blowing agent a particle size of 325-400 mesh size, antistatic and flame retardant in thermoset a particle size of 325 mesh size.

Preferably, the carbonised vegetative-based filler is carbonised rice husk.

Preferably, the carbonised rice husk is burnt at about 800°C for about 4 seconds. Most preferably, the carbonised rice husk is burnt at 803-804°C for 3-4 seconds.

In another aspect the invention resides in a composite material, more particularly a composite plastic including a vegetative-based filler when used as a conductive or flame retardant article.

Preferably, the carbonised vegetative-based filler is carbonised rice husk which has been burnt at 803-804°C for 3-4 seconds.

The present invention also exhibits the usage of fresh and carbonised rice husk as a blowing agent when used in combination with recycled (reclaimed), or virgin natural rubber or other suitable thermoplastic materials. Though other conventional blowing agents have been used with natural or synthetic rubber to achieve the similar products but so far no blowing agents have been used with recycle (reclaim) rubber to produce similar products. Furthermore the conventional blowing agents are expensive and dosages are higher as compared to the fresh rice husk. For example for the conventional blowing agent, the dosage is about 6.5-7 phr, whereas the fresh/carbonised rice husk, the dosage is between 1.5 to 3 phr. When rice husk is used at different dosages the blowing effect is different. It was also noted that the rice husk does not only work as a

blowing agent, but also as a plasticizer and a filler. The properties achieved are comparable to conventional blowing agents, when using fresh or carbonised rice, has no difference to the conventional blowing agent other than the colour of the end product.

5 Ebonite, a hard, dark-coloured plastic-like material, is the reaction product of rubber and a large proportion of sulphur. Simple rubber / sulphur mixtures are seldom used in practice; they suffer from poor processability, require long cure times and lead to excessive shrinkage and heat evolution during cure. Accelerators, fillers, processing aids and other compounding ingredients are
10 widely used in ebonite, as in soft rubber vulcanised rubber, to ease processing, shorten cure times and modify properties. The curing times for ebonite are generally up to ten (10) hours at 150° C, thus making ebonite products expensive. Ebonite can be made from synthetic, such as BR, NBR, SBR and Nitrile rubber and as well as from Natural rubber. High strength, low thermal conductivity,
15 chemical resistance and insulating properties of natural rubber make it a popular choice. Although it has been superseded in many applications by synthetic thermoplastic and thermosets, it is still used for outstanding chemical resistance and electrical properties coupled with high mechanical strength and ease of machining.

20 The present invention exhibits the usage of fresh and carbonised rice as an accelerator when used in combination with recycled (devulcanised) or virgin natural rubber, and at the same time making ebonite a conductive product when carbonised rice husk is used. Though other conventional accelerators have been used with natural or synthetic rubber (virgin or recycled) to achieve the similar
25 products but so far no accelerators like the rice husk material have been used with recycled (devulcanised) rubber to produce similar products. Further more the conventional accelerators and conductive carbon black are expensive and difficult to blend and process. When rice husk is used singularly at different dosages the activation effect is different to meet industrial requirements.
30 Generally for ebonite production the sulphur content should be in the range of 25-40 phr, but whereas when fresh rice husk between 25-30 phr is used the sulphur content could be reduced to 20-25 phr. Accelerators are less effective in ebonite than in soft rubber and large quantities are generally required. Basic accelerators

such as guanidines and aldehyde-amines are preferred. Inorganic activators such as magnesium oxide, magnesium carbonate and lime are also effective when used with organic accelerators to reduce cure time without the risk of over heating.

5 Common inorganic fillers used in ebonite are china clay, talc, silica, whiting and magnesium oxide. These also reduce shrinkage and heat evolution but loaded ebonite generally have weaker mechanical properties than unloaded ones. Carbon black does not reinforce ebonite and is normally only added for pigmentation, although conductive carbon black are sometimes used to prepare
10 electrically conducting ebonite.

BEST MODE

Following is an example of the invention, in this example the filler is carbonised rice husk (CRH) which has been burnt at 803-804°C for 3-4 seconds.
15 After this the CRH is obtained.

The manner in which the rice husk is burnt is believed to be important in achieving the desired surface area, surface structure and porosity necessary for conductivity and flame retardant and blowing properties in the composite plastics to be achieved. At this stage the range of temperature and the duration of the
20 time of burning which achieves the desired result has not been fully explored, however it is predicted that the temperature range will be from about 100-950°C and the time range will be from about 2-30 seconds, although these ranges may be much narrower. The importance of controlled burning in a prescribed time results in obtaining better surface area and porosity which in turn offers ideal
25 properties emitting anti-static, flame retardant and enhancing physical properties of the material. In the absence of controlled burning, the surface area, surface structure and porosity would be distorted. While the present exemplification involves use of carbonised rice husk it is possible that the desired results may be achieved by use of other carbonised vegetative-based fillers.

30 EXAMPLE 1

A thermoset application called pulforming was used to manufacture fibre reinforced bolts. Fibre glass tows (36 tow of 8000 tex) are pulled into a resin bath that contains:

1. Polyester and Vinyl Ester combination, ie. 60% Vinyl Ester (Derakane 411 - Dow Chemical) and 40% Polyester (Everpol 3260 AR - P.T. Arinde).
2. Zinc Stearate (mould releasing agent) - 1.18% of the resin weight
BYK 980 (improves wetting and dispersing of fillers in glass fibre reinforcement compounds) - 1.5% of the filler weight.
3. BYK 515 (air releasing agent) - 0.5% of the total weight of the resin mixture.
4. BYK 996 (wetting and dispersing additive for mineral fillers in hot curing, glass fibre reinforced UP-resin systems) - 2% of the resin weight.
5. Fillers ($\text{Ca}_2(\text{CO})_3$ & Carbonised Rice Husk (mesh size 325) @ 55% & 12% of the resin weight).
6. Aluminium Trihydrate (2.4% of the resin weight).
7. Catalyst TBPH (Tertiary Butyl Peroxy-2-Ethyl Hexanoate)
- 2.12% of the resin weight.
TBPB (Tertiary Butyl Perbenzoate)
- 0.53% of the resin weight.

The wet fibre glass tow is pulled into the mould and compressed at a pressure of 800psi (5600kPa) for 3.8 minutes at 130°C. Then the bolt is pulled out of the mould and left to cure.

The following day tests were carried out on the bolt with the following results:

Tensile strength at the thread - 50kN

Torque - 45ft/lb

Bond strength - BS 1610:Part 1, Grade 1.0

Fire rating - BS 5865:1980 - Persistence flame shall be less than 10 seconds

Electrical conductivity - less than 10 to the power of 9 Ohms.

EXAMPLE 2

All chemicals used are taken by percentage of weight of rubber. The rubber and the chemicals are mixed in a Banbury, for 5 minutes. Recycled rubber

(reclaim) (220g) is first milled with zinc oxide (4.5%) - accelerator, which is followed with stearic acid (1.8%) - activator, Mercadibenzothiazole disulphide (MBTS)(0.5%), Tetramethylthiuram disulphide (TMTD)(0.2%)- accelerator, fresh rice husk (27%)-blowing agent and filler and sulphur (2.7%)- vulcanisate. Then the mixed compound is milled for five (5) minutes to form a sheet that is ready for curing. Then a piece of the sheet weighing about 32g is placed in a mould that it is to be cured for two (2) minutes in a oven at 150°C temperature. The conventional curing time is six (6) minutes at the same temperature of 150°C.

The rubber and the chemicals are mixed in a Banbury, for 5 minutes. The similar approach has been done for using SBR Rubber (100g), silica (62g), Peg 1500 (2.5g), Paraffin oil (5g), Zinc oxide (2.5g), Wing stay (0.5g), Wax (1g), Mercadibenzothiazole disulphide (MBTS) (1.5g), Tetramethylthiuram disulphide (TMTD) (0.2g), Stearic acid (1.5g) and Sulphur (2g). The milling was done for ten (10) minutes and later cured in the oven for six (6) minutes at 150°C.

This exercise was repeated by using fifty (50) percent of the virgin material compound and fifty (50) percent recycled (reclaimed) material compound, and cured in the oven at 150°C for two (2) minutes.

With the level, of rice husk dosage, the blowing effect can be controlled to suit the industry's requirement.

Machine Operating Conditions

Virgin Rubber:

Mix properties; White filled mix

Mooney viscosity, MLI + 3, 100°C	24
Mooney viscosity, MLI + 3, 120°C	18.5
Mooney scorch, t ₅ MLI + 3, 120°C min	5.8
Monsanto Rheometer, 160°C	
time to 95% cross-linking, s	110

By using rice husk the Mooney viscosity was lower than the conventional filler, thus lowering scotch time (time taken by the rubber compound to flow into the

mould) and curing time (time taken to cure rubber compound) respectively. As such this leads to a cheaper production system. Presently various fillers and blowing agents are being used in the production of soft/spongy rubber that would produce different types of cell structures for an end product, but the cost
5 determines the market.

EXAMPLE 3

All chemicals used are taken by percentage of weight of rubber. The rubber and the chemicals are mixed in a Banbury, for six (6) minutes. The
10 recycle (devulcanised) rubber is first milled with magnesium oxide (2%) - accelerator, which is followed with Diphenylguanidine (2%) - accelerator, fresh rice husk (30%) - accelerator and filler and sulphur (30%) - vulcanisate. After the milling at the Banbury for ten (10) minutes, it is then milled into a sheet. The mould was heated in the oven press to 150°C then the sheeted rubber is placed
15 in the mould and it is cured for twelve (12) minutes. The conventional curing time is between eight to ten hours at the same temperature of 150°C.

A conventional formula for ebonite was selected to compare. The rubber and the chemicals are mixed in a Banbury, for 5 minutes. The similar mixing as above was followed, using SBR 5 Rubber (100g), ebonite dust (100g), China clay
20 (50g), Magnesium oxide (5g), Diphenylguanidine (3g), Linseed oil (5g) and Sulphur (45g). The milling was done for ten (10) minutes and later cured in the oven for eight (8) hours at 150°C.

Mix properties; Rice Husk filled mix

25	Mooney viscosity, MLI + 3, 100°C	24
	Mooney viscosity, MLI + 3, 120°C	18.5
	Mooney scorch, t ₅ , MLI + 3, 120°C min.	5.8
	Monsanto Rheometer, 160°C	
	time to 95% cross-linking, s	110

By using rice husk the curing time is reduced tremendously twelve minutes as compared to eight to ten hours. The sulphur content in the rubber polymer is reduced by fifteen percent.

5 EXAMPLE 4

All chemicals used are taken by percentage of weight of natural rubber (NR). The natural rubber and the chemicals are mixed in an open mill or kinder, for six (6) minutes. Natural rubber is first milled with stearic acid (1%) and zinc oxide (5%) activator, which is followed with rice husk (blowing agent) (2.5-3.5%), calcium carbonate - (40%), promoter - urea based (2.5-3.5%), silica (10%), accelerator dibenzthiazyldisulphide (MBTS) (0.05%) and catalyst sulphur (1.5%). After the milling at the open mill or kinder for ten (10) minutes, it is then milled into a sheet. The mould was heated in the oven press to 160°C then the sheeted natural rubber is placed in the mould and it is cured for twenty-two (22) minutes.

The temperature for curing could be from 145°-160°C and the cure time may differ according to the mould size.

Cured properties; rice husk filled blowed mix - micro-cellular cells.

1.	Hardness	Askar C	35
20 2.	Shrinkage	%	5
3.	Specific Gravity	g/cc	0.3-0.35

By using rice husk as a blowing agent the catalyst percentage could be reduced and as well as the percentage of blowing agent used.

25

EXAMPLE 5

THERMOPLASTIC (EVA)

All chemicals used are taken by percentage of weight of thermoplastic - Ethyl Vinyl Acetate (EVA). The EVA and the chemicals are mixed in an Open Mill or Knider, for six (6) minutes. Ethyl Vinyl Acetate (EVA) is first milled with Stearic Acid (1%) and zinc oxide (5%) accelerator, which is followed with Rice Husk (Blowing agent) (2.5%). Magnesium carbonate - (10%), Promoter - urea based (5%) and catalyst Diacyl Peroxide (1%). After the milling at the Open mill or

Knider for ten (10) minutes, it is then milled into a sheet. The mould was heated in the oven press to 160°C then the sheeted EVA is placed in the mould and it is cured for twenty-two (22) minutes.

5 The temperature for curing could be from 145°-160°C and the cure time may differ according to the mould size.

Cured properties: Rice Husk filled blowed mix - micro-cellular cells

1.	HARDNESS	Askar C	29-35
2.	SHRINKAGE	%	2
3.	SPECIFIC GRAVITY	g/cc	0.200
10 4.	COMPRESSION SET	%	80

By using rice husk as a blowing agent the catalyst percentage could be reduced and as well as the percentage of blowing agent used.

15 **EXAMPLE 6**

The rice husk is mixed by weight with tyre crumbs(35-40 mesh) and an effluent from the palm oil mill called Scavenger (which have a fatty acid content (C₈-C₁₈). From literature it has been reported that by using tyre crumb with the binder (bitumen) there is an improve of properties for the asphalt mixture. This invention not only uses tyre crumb along with rice husk and an oil palm effluent to further improve the properties. As well as the formulation address the recyclability of all agro waste by-products to be used in the road surfacing industries. The formulation of the rice husk mixture as follows:

25	Rice husk	50%
	Tyre crumb	45%
	Scavenger	5%

In this particular example the usage of rice husk mixture is divided into two categories:

- 30 A. RICE HUSK MIXTURE USED IN MODIFIED BINDER
 B. RICE HUSK MIXTURE USED IN AS FILLER

- 5 A. The rice husk mixture is added to the bitumen first in compliance to the SOCIETY OF HIGHWAY PROCEDURE (SHRP) to manufacture modified bitumen. The bitumen is first heated to about 160°C, then the rice husk mixture twenty percent 20% by weight of bitumen is mixed with the heated bitumen for about one hour. As a result of this reaction a modified bitumen is made. From here 5-7% by weight of this modified bitumen is added to the aggregate. The aggregate is first heated to about 200°C and the modified bitumen is mixed for three to four minutes. The modified bitumen with rice husk mixture complies to all requirement of the SHRP.
- 10 B. The rice husk mixture is added as a filler to the aggregate, by four (4%) by weight to the aggregate weight. The aggregate is first heated to 200°C, and is allowed to cool to about 160°C, then the rice husk mixture is added and mixed and lastly the bitumen 5-6% by weight of aggregate is added and mixed for 3-4 minutes. This blending with rice husk mixture complies
- 15 to all requirement of the Marshall Properties.

TABLE 1

PROPERTIES OF RICE HUSK MODIFIED BINDER - SHRP

SHRP	80/100	RICE HUSK MIXTURE
FLASH POINT TEMP. °C	230	240
SOFTENING POINT, °C	44-50	55-70
PENETRATION @ °C 25, dmm	80-100	60-100
BROOKFIELD VISCOSITY @ 135°C, MPaS	<500	>1500
DYNAMIC SHEAR RHEOMETER		
PG 70		
ORIGINAL		
G* (Pa)	<1000	>1200
$\delta(^{\circ})$	>80	>80
G*/Sin δ	<1000	>1200
AFTER RTFOT		
G* (Pa)	<1000	>3800
$\delta(^{\circ})$	>80	>70
G*/Sin δ	<1000	>3800
AFTER PAV		
G* (Pa)	<1000	>230
$\delta(^{\circ})$	>80	>50
G*/Sin δ	<1000	>260
PG 76		
ORIGINAL		
G* (Pa)	<1000	>1800
$\delta(^{\circ})$	>80	>70
G*/Sin δ	<1000	>1800
AFTER RTFOT		
G* (Pa)	<1000	>2400
$\delta(^{\circ})$	>80	>70
G*/Sin δ	<1000	>2600
AFTER PAV		
G* (Pa)	<1000	>230
$\delta(^{\circ})$	>80	>50
G*/Sin δ	<1000	>280

TABLE 2

MIXED PROPERTIES OF RICE HUSK MODIFIED BINDER

PROPERTIES	80/100	RICE HUSK MIXTURE
MARSHALL STABILITY (kN)	5-10	>13
FLOW (mm)	2-4	2-4
QUOTIENT (kN/mm)	1-3.5	3-4
RESILIENT MODULUS @ 25°	>2000	>2800

TABLE 3

MIXED PROPERTIES OF RICE HUSK MIXTURE AS FILLER

PROPERTIES	80/100	RICE HUSK MIXTURE
MARSHALL STABILITY (kN)	6-10	>12
FLOW (mm)	2-4	2-4
QUOTIENT (kN/mm)	1-3.5	3-4
RESILIENT MODULUS @ 25°	>2000	>2800

TABLE 4

PREFERRED PARTICLE SIZE AND DOSAGE OF FRESH AND/OR CARBONISED RICE HUSK FOR PARTICULAR COMPOSITE MATERIALS

COMPOSITE MATERIAL	FRESH/DOSAGE	CARBONISED/DOSAGE
BITUMEN (MECHANICAL PROPERTY)	100 MESH - 40-60 phr	-
THERMOPLASTIC (EVA) BLOWING CHARACTER	325-400 MESH - 1.5-2.5 phr	325-400 MESH - 1.5-2.6 phr
THERMOPLASTIC (EVA) MECHANICAL PROPERTY	325-400 MESH - 1.5-5 phr	325-400 MESH - 1.5-2.5 phr
RUBBER (N.R./S.R.) BLOWING CHARACTER	325-400 MESH - 1.5-27 phr	325-400 MESH - 1.5-27 phr
EBONITE N.R. (REDUCE CURE TIME)	100-200 MESH 18-30 phr	-
RUBBER (N.R./S.R.) SCOTCH TIME	100-200 MESH 5-10 phr	100-200 MESH 5-10 phr
THERMOSET RESIN (FLAME PROPERTY)	-	325 MESH 10-15 phr
THERMOSET RESIN (MECHANICAL PROPERTY)	100-200 MESH 10-15 phr	100-200 MESH 10-15 phr
THERMOSET RESIN (ANTISTATIC)	-	325 MESH 10-15 phr
RUBBER-LATEX (N.R./S.R.) ANTISTATIC	-	325-400 MESH 5-15 phr
RUBBER (N.R./S.R.) ANTISTATIC	-	325-400 MESH 5-15 phr
CONCRETE (MECHANICAL PROPERTY)	100-200 MESH 10-15 phr	100-200 MESH 10-15 phr

N.R. - NATURAL RUBBER
S.R. - SYNTHETIC RUBBER

10 It will of course be realised that whilst the above has been given by way of illustrative examples of this invention, all such and other modifications and variations hereto, as would be apparent to persons skilled in the art, are deemed to fall within the broad scope and ambit of this invention as herein set forth. For instance, while the preceding examples relate to the use of fresh and/or
15 carbonised rice husk it would be apparent to a person skilled in the art that other cereal husks such as sorghum husk may be suitable.

Throughout the description and claims of the specification where reference is made to the dosage of fresh and/or carbonised rice husk, this dosage is expressed in terms of "phr" (parts per hundred) based on the weight of the composite material into which the rice husk is being introduced.

- 5 Throughout the description and claims of the specification the word "comprise" and variations of the word, such as "comprising" and "comprises", is not intended to exclude other additives, components, integers or steps.

CLAIMS:

1. A filler for use in composite materials wherein said filler comprises a vegetative-based material and wherein said vegetative-based material can be fresh or carbonised.
2. A filler according to claim 1 wherein said vegetative-based material comprises cereal husk.
3. A filler according to claim 2 wherein said cereal husk comprises carbonised cereal husk.
4. A filler according to claim 3 wherein said carbonised cereal husk comprises carbonised rice husk.
5. A filler according to claim 4 wherein said carbonised rice husk is the product of burning fresh rice husk at about 800°C.
6. A filler according to claim 5 wherein said carbonised rice husk is the product of burning fresh rice husk at about 800°C for about 4 seconds.
7. A filler according to claim 6 wherein the carbonised rice husk is the product of burning fresh rice husk at 803 to 804°C for 3 to 4 seconds.
8. A process for the production of a carbonised vegetative-based filler wherein said process comprises burning a fresh vegetative-based material at about 800°C for about 4 seconds.
9. A process according to claim 8 wherein the fresh vegetative material is ground to a particle size of from 100 mesh to 400 mesh.

10. A process according to claim 8 or claim 9 wherein said fresh vegetative-based material is burned at about 803 to 804°C for 3 to 4 seconds.

11. A process according to any one of claims 8 to 10 wherein said process
5 utilises fresh rice husk as the vegetative material.

12. A process according to claim 11 wherein the fresh cereal husk comprises fresh rice husk.

10 13. A filler according to any one of claims 1 to 7 when produced by a process according to any one of claims 8 to 12.

14. A method for improving the anti-static, flame retardant, accelerator, plasticiser and/or blowing characteristics of a composite material wherein said
15 method comprises blending into the composite material with a carbonised vegetative-based filler according to any one of claims 1 to 7 and wherein said blending is substantially completed prior to incorporation of any additives, if any.

15. A method according to claim 14 wherein the carbonised vegetative filler
20 has a particle size of from 100 mesh to 400 mesh.

16. A method according to claim 14 or claim 15 wherein the composite material is latex (NR/SR) the dosage of the carbonised vegetative filler is from 1.5 to 2.5 phr (parts per hundred).

25 17. A method according to any one of claims 14 to 16 wherein said composite material is selected from the group comprising:

- i) thermoplastic resins;
- ii) thermoset plastics;
- 30 iii) rubbers and elastomeric materials;
- iv) conductive coatings;
- v) printing inks;
- vi) bitumen; and

vii) concrete.

18. A composite material having improved anti-static, flame retardant, accelerator, plasticiser and/or blowing characteristics wherein said composite material is produced by the method of any one of claims 14 to 17.

19. A method for improving the mechanical properties of bitumen, said method comprising blending fresh rice husk into said bitumen.

20. A method according to claim 19 wherein the rice husk has a particle size of from 100 to 200 mesh and the dosage of rice husk is between 40 to 60 phr.

21. A method for improving the blowing character of a thermoplastic resin this method comprising blending fresh and/or carbonised rice husk into said thermoplastic resin.

22. A method according to claim 21 wherein the rice husk has a particle size of between 325 to 400 mesh and the dosage of the rice husk is between 1.5 to 2.5 phr.

23. A method for improving the mechanical properties of thermoplastic resin including compression strength, said method comprising blending rice husk into said thermoplastic resin.

24. A method according to claim 23 wherein the rice husk as a particle size of between 325 to 400 mesh and the dosage of the rice husk is between 1.5 and 2.5 phr.

25. A method for improving the blowing character of rubber said method comprising blending fresh and/or carbonised rice husk into said rubber.

26. A method according to claim 25 wherein the rice husk has a particle size of between 325 to 400 mesh and the dosage of the rice husk is between 1.5 and 27 phr.

5 27. A method for reducing the cure time of ebonite NR wherein said method comprises blending fresh rice husk into said ebonite NR.

28. A method according to claim 27 wherein the rice husk has a particle size of between 100-200 mesh and the dosage of the rice husk is between 16 to 30 phr.

10

29. A method for improving the scotch time of rubber said method comprising blending fresh and/or carbonised rice husk into said rubber.

30. A method according to claim 29 wherein the rice husk has a particle size of
15 between 100 to 200 mesh and the dosage of rice husk is between 5 to 10 phr.

31. A method for improving the flame retardant ability of a thermoset resin said method comprising blending carbonised rice husk into said thermoset resin.

20 32. A method according to claim 31 wherein the carbonised rice husk has a particle size of 325 to 400 mesh and the dosage of carbonised rice husk is between 10 to 15 phr.

25 33. A method for improving the mechanical properties of thermoset resins including tensile and torque strength, said method comprising blending fresh and/or carbonised rice husk into said thermoset resin.

34. A method according to claim 33 wherein the rice husk has a particle size of between 100 to 200 mesh and the dosage of rice husk is between 10 to 15 phr.

30

35. A method for improving the antistatic properties of a thermoset resin said method comprising blending carbonised rice husk into said thermoset resin.

36. A method according to claim 34, wherein the carbonised rice husk has a particle size of between 325 to 400 mesh and the dosage of carbonised rice husk is between 10 to 15 phr.

5 37. A method for improving the antistatic properties of rubber and/or rubber-latex said method comprising blending carbonised rice husk into said rubber and/or rubber-latex.

10 38. A method according to claim 37 wherein the carbonised rice husk has a particle size of between 325 to 400 mesh and the dosage of carbonised rice husk is between 5 to 15 phr.

39. A method for improving the mechanical properties of concrete said method comprising blending fresh and/or carbonised rice husk into said concrete.

15

40. A method according to claim 39 wherein the rice husk has a particle size of between 100 to 200 mesh and the dosage of the rice husk is between 10 to 15 phr.

20

41. A filler according to claim 1 substantially as hereinbefore described with reference to any of the examples.

42. A process according to claim 8 substantially as hereinbefore described with reference to any of the examples.

25

43. A method according to claim 14 substantially as hereinbefore described with reference to any of the examples.

30

44. A composite material according to claim 18 substantially as hereinbefore described with reference to any of the examples.